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VOL. 37 No. 332 AUGUST 1951

THIRD CADMAN MEMORIAL LECTURE*

COMPETITIVE AND CO-OPERATIVE RESEARCH IN THE AMERICAN PETROLEUM INDUSTRY

By ROBERT E. WILSON †

INTRODUCTION

I AM highly appreciative of the great honour you have done me in inviting me to deliver the third Memorial Lecture in honour of Lord Cadman, that great representative of your country and our industry. It is also a high honour to speak in this Royal Institution, which has reverberated for a century and a half to the voices of so many illustrious scientists. It is inspiring to recall these great pioneers of chemistry and physics who contributed so much to my chosen field of chemical engineering, and particularly Faraday, who initiated hydrocarbon chemistry more than a century ago in this very building by separating benzene and determining its composition as well as that of ethylene and butylene.

My being here is not due to any present or even past eminence as a scientist. Nor am I the proper individual to elaborate upon, or to try to project into the future, the fine accounts of research in our industry which were presented by my two predecessor Cadman lecturers.

In an attempt to offer you a petroleum research topic on which I might speak with some authority twenty years after leaving the laboratory, I shall discuss the organization and philosophy of research in America —and particularly the pros and cons of competitive versus co-operative research, in the light of our experience. This does not mean that I believe that the way we carry out research in the United States is the only way. It is simply a way that has worked well for us over the years.

SOME CONTRIBUTIONS OF PETROLEUM RESEARCH

Just thirty-five years ago this month I was graduated from the Massachusetts Institute of Technology and became interested in research in oil refining. Let me recall a few of the amazing developments since then in our industry which can be attributed largely to its research activities:

(1) The actual yield of gasoline from a barrel of crude oil has more than doubled, and the potential yield has quadrupled. That potential yield could and would be attained except for the growing demand for so many other products from petroleum.

(2) The average octane rating of motor gasoline has been increased from 50 to 87. This means that, in an engine designed to utilize it

* Delivered in the Lecture Hall of the Royal Institution on June 21, 1951.

† Chairman of the Board, Standard Oil Co. (Indiana).

effectively, *one gallon of to-day's gasoline will do the work of two of 1916 quality*.

(3) To-day's piston engine for military aircraft will produce over ten times as much power, with half the weight per horse-power, as the best engines in the first world war. While engine research and metallurgical research have helped substantially, the tremendous improvements in the quality of aviation gasolines and lubricating oils have been the major factors in this result.

(4) In spite of all the improvement in quality, the price of gasoline (excluding taxes) since the second world war has been so low that it represents a saving to the United States public of over *four billion dollars* per year as compared with the 1920 price! Relatively speaking, gasoline is the cheapest major product on the market to-day. Technology has played a major part in keeping costs comparatively low, and hence in stimulating the sixfold expansion of the American industry since 1916.

(5) Over 90 per cent of the molecules in to-day's military aviation gasoline are not present in the crude oil, but are made by synthetic chemical processes, most of which were unknown, even to scientists, twenty years ago. Even our motor gasoline contains about 60 per cent of synthetic molecules. Research has transformed petroleum refining into essentially a chemical-synthesis industry. The magnitude of its operations dwarf all the synthetic processes and products of the "old line" chemical industry. Consider a few examples:

Organic Chemical Industry		Synthetics from Petroleum	
Product	Millions of pounds per year	Product	Millions of pounds per year
All plastics . .	1,700	Alkylate . . .	16,000
Detergents . .	330	Alcohols . . .	2,500
Nylon . . .	170	Rubber . . .	1,200

In the last twenty-five years the proportion of our total organic chemicals produced from petroleum has increased from less than a tenth of 1 per cent to about 35 per cent!

(6) The benefits of applied research to the industry and to the public have been so outstanding that the number of technically trained men engaged in research and development in our industry is at least forty times as large as in 1916. The annual expenditures are now in excess of $100 million per year. While cost is a poor way of measuring the value of research, it does demonstrate management's confidence in its past and prospective worth.

How Did It Happen?

Both the growth of applied research in our industry and its achievements during the past third of a century are unprecedented phenomena which warrant analysis as to how it all happened. Yes, "happened" is the word, because it was a gradual evolution and not the result of any long-range plan.

About 90 per cent of our petroleum research is carried on competitively in private laboratories. This active competition between industry laboratories has been largely responsible for the development of dozens of novel and complicated processes. These include, in the field of motor fuel alone, different methods of polymerization, isomerization, hydroforming, desulphurization, extractive distillation, and thermal and catalytic cracking. Moreover, every process has been made more efficient with the passing years, as each operator has tried to gain a little advantage over his competitors in the economic race.

Competitive *v.* Co-operative Research

There are still a few who argue that competitive research involves too much duplication of effort and waste of scientific manpower. They urge that the government or trade associations should carry out most applied research on a co-operative basis in the expectation that this would reduce overall costs. But reduction in research costs should certainly not be the main objective—the real question is, what would happen to our rate of progress under such conditions?

Co-operative research suffers from the absence of the incentive to make a profit, or to get ahead in the competitive struggle. It also abandons the proved advantages of many independent centres of initiative and different methods of attack on difficult problems. Furthermore, if duplication is really to be avoided, there would have to be centralized control, with each research worker assigned to a specific problem, regardless of his individual interests or desires.

It is hardly necessary to say to this group that no individual, board, or bureaucratic organization is wise enough to effectively administer any such set-up. During the war Germany had centralized control of research, and it failed signally to make good use of its outstanding scientific manpower, once the dictatorship attained full control. By all logic Germany, with so many of the world's best scientists, should have beaten the Allies to the atomic bomb. Indeed, worry about this very possibility greatly stimulated competitive research efforts in both Britain and the United States. The post-war analysis of German wartime research showed convincingly that centralized control was its greatest weakness, especially in fields requiring real imagination and initiative. Another sad example is the research being done by the Lysenko group in Russia—if the frantic hunt for evidence to support a false theory, dictated from above, can properly be called research.

However, even the strong advocates of competitive research realize that such competition can be carried to extremes. If there were general unwillingness to publish results promptly, once they are protected by patent applications, or to disclose and license new processes as soon as they are developed, scientific and industrial progress would be slowed down. As Kettering well said, whoever locks his laboratory door locks out much more than he locks in. Fortunately, our patent system encourages prompt disclosure and interchange of information, without jeopardizing the inventor's rights to his discovery.

It must also be conceded that there are some types of research which can be effectively carried out only by co-operative effort. One outstanding

example in the United States is the Co-ordinating Research Council, which has been set up by the automobile and petroleum industries to work on problems of adapting engines to fuels and lubricants, and vice versa. The Council has contributed greatly to progress and better understanding between the two industries. In some fields, like standardization, industrial safety, and the development of testing methods, wide co-operation is essential to success. And in the important field of developing basic information and fundamental data, the co-operative effort of a whole industry is sometimes the only way such work can be adequately financed.

However, it is difficult to make clear and convincing the points I have in mind by discussing research in the abstract. The concrete example of what actually happened in connexion with the development of cracking in the American oil industry illustrates many of these points; and it has been duplicated in principle, though over a shorter time period, in many of the later process developments of our industry.

The Development of Cracking

The development of cracking furnished the main driving force for the rapid expansion of American research activities after the first world war. I shall review it, not from the technical viewpoint, but to analyse the incentives which were effective and the far-reaching results, both direct and indirect.

Our industry had been in existence for thirty years before it made any appreciable attempt towards research on anything. There were several reasons for this. First, the chief competing industries were not exactly stiff competition. Whale oil and coal oil were inferior to kerosine as burning oils, and were much more expensive. Pennsylvania crude was for many years easy to find, cheap to develop, and very easy to refine. Where there are few problems there is little research.

So, it was not until 1889 that the first highly trained chemist was hired in the petroleum industry. He was Dr William M. Burton, just out of Johns Hopkins University. Mr John D. Rockefeller personally directed his being hired to help Standard Oil solve the problem of making marketable products from the recently discovered " sour " crude oil from Lima, Ohio. As usual it took a tough problem to get research started. Dr Burton fitted up a pioneer laboratory in two bedrooms of an old farm-house at Whiting, Indiana.

But not until twenty years later—a full half-century after the start of the industry—did he initiate his pioneer work on cracking. By that time he was plant superintendent, and became concerned about the need for some method of greatly increasing gasoline yields. If gasoline production were to be restricted to the 15 or 20 per cent which could be separated from average crude oil by fractionation, the price would soon go so high as to stifle the development of the automobile industry—or else divert it to using steam or electric storage batteries, rather than gasoline.

Of course, if some of our modern bureaucrats had been in control, the problem would have been handled very simply, merely by slapping a price ceiling on gasoline, and then rationing it as it became scarce! We might never have had a cracking process—and the United States would not to-day be a nation on wheels.

Not having to worry, however, about such interference with the laws of supply and demand, Dr Burton, with the real spirit of the entrepreneur, decided that the whole Whiting research staff should be put on this problem. While this staff was probably the best and largest in the industry, it actually consisted only of Dr Robert E. Humphreys and his assistant, Dr F. M. Rogers, two other Johns Hopkins men. After two years of travail, which I shall not take the time to detail, a successful cracking process was developed, and the first commercial Burton stills went into operation in the Whiting (Indiana) refinery in January 1913.

The process was perfected in the nick of time, and was a major factor in helping to meet the heavy gasoline demand during the first world war. For more than seven years it, with some improvements, was the only cracking process in real commercial operation. During that period Standard of Indiana licensed fifteen other companies, including many of the other Standard Oil companies from which it had recently been divorced by the 1911 dissolution decree. Its royalties during this period aggregated about $15,000,000 though they were set at a level approximating only 10 or 15 per cent of the profits of the operation.

Results of the Cracking Development

The new process had many direct and indirect results. It doubled the yield of gasoline obtainable from crude, and so assured the future of the gasoline automobile. It showed the industry that it need not confine itself to separating and purifying what came from the ground, but could change the size and shape of the naturally occurring molecules. The comparatively high temperatures and pressures employed created many related technological problems and stimulated research in the fields of metallurgy and refinery engineering. The by-products of cracking included large quantities of reactive olefinic gases, and greatly stimulated research by chemical companies such as Carbide and Carbon Chemicals, du Pont, and Imperial Chemical Industries, all of whom sought ways of using these interesting low-cost raw materials. It brought technical men into the industry, and they soon penetrated every branch, stimulating more research wherever they went. They even infiltrated into boards of directors! To-day my own board of eleven includes nine " inside " directors, of whom three were trained as chemical engineers, two as mechanical engineers, one as a mining engineer, and one as a Ph.D. chemist!

Stimuli to Competitive Research

The growing amount of royalties, whether received or paid, constituted a tremendous stimulus to the competing companies to do research on cracking. Standard of Indiana was by far the largest oil company operating in the Middle West. The other companies operating in that area did not like to aid their principal competitor by paying royalties to it. To paraphrase a famous remark made in 1798 by an American diplomat, some of Standard's competitors believed in " millions for research, but not one cent for tribute." Once Dr Burton had shown that cracking could be made commercial, the urge to do research on the process became almost universal.

Another incentive arose from the belief in some quarters that Standard's

early patents were of questionable validity, or that they should be construed as being restricted to an operation wherein the pressure in the cracking zone extended through the condensing system. This led some companies to conclude that research of their own might enable them to get around these patents, or possibly establish that the patents were not valid over certain prior art teachings, such as, those of your Sir James Dewar and Sir Boverton Redwood.

Litigation as a Factor in Research

The legal problems arising from these doubts and uncertainties were themselves an incentive to research. This is not uncommon. Patents are generally taken out on the pioneering fringe of technology, where information as to what really happens is seldom complete. Litigation is often necessary to establish the true inventor of an important new process or product. And while research often leads to litigation, it is also true that litigation often leads to research. Certainly this was the case in the early history of cracking. In order to obtain information needed in the prosecution of patent suits, or in setting up a defence against such suits, companies carried out extensive research, which resulted in much better understanding of, and major improvements in, the cracking art.

I must admit that still another stimulus to research by our competitors was the fact that Standard of Indiana rather tended to rest on its oars, instead of pushing vigorously the additional research necessary to make major improvements in the cracking process, though it did develop the Clark tube still and other substantial betterments. While the Texas Company, Universal Oil Products, and several other companies were working on attempts to make cracking continuous, Standard's experience with the rapid coking of all surfaces in contact with the oil being cracked, and the apparent impracticability of pumping oil at anything like cracking temperatures, convinced our people that a continuous cracking process was not feasible. As a chemical engineer, I believe one reason for this attitude was that the Whiting staff consisted almost entirely of chemists, who lacked the basic conviction held by chemical engineers that continuous processes are so superior that they are bound to replace batch processes, even though the reactions are essentially the same.

In any case, the value of competitive research soon showed up. Within a few years other laboratories, which did not share Standard's pessimism about continuous cracking, worked out several different successful methods of accomplishing it. As a matter of fact, it fell to my lot soon after my arrival at Whiting in 1922 to tell Dr Burton and Dr Humphreys that the Burton process was obsolete. Fortunately, they received the information as true scientists would be expected to, and after several years of lost time Whiting got back into the active development of still better cracking processes.

Competitive Developments in Cracking

I can mention in passing only a few of the achievements of all this competitive research on cracking. Trumble alleviated the coke problem by bleeding off part of the heavy cracked residue from a cyclic system. Thomp-

son developed the clever device of a surge pump, which made it possible to pump hot oil and gave great impetus to continuous operation. This device was later succeeded by the centrifugal hot-oil pump. The concept of clean circulation was developed by Behimer, and was embodied in the later modifications of the Holmes–Manley units widely used and licensed by the Texas Company. Ellis, and other inventors working with the Standard Oil Company of New Jersey, developed the tube and tank process. In this a pipe coil was used for heating the oil to cracking temperature, but the bulk of the cracking and coke deposition took place in an unheated soaking drum. An important development was achieved by the Cross brothers, who recognized the desirability of using very high pressures to improve heat transfer and minimize coke formation. The Cross brothers' endeavours were underwritten by a group of individuals not connected with the industry. One of them was a manufacturer of hairpins! Universal Oil Products Company developed the Dubbs cracking process, which made early use of the clean circulation feature, and also developed the non-residuum or coking type of operation. The Universal venture was mainly financed by a Chicago meat packer, Mr J. Ogden Armour. There were many other less important or less successful cracking process developments both within and without the petroleum industry.

It will be noted that this research was carried out by small companies as well as large. The equipment required for this early work was not expensive. Even the commercial units themselves were relatively small, and much experimentation was possible on the full-scale refinery equipment. Even if a new commercial unit did not represent as great an improvement over its predecessors as had been hoped, it was usually a good operable unit, which made gasoline and paid its way.

There was then no need for long and costly pilot-plant work. Commercial stills cost less than some of to-day's pilot plants! Refiners did not have to take the chance, as we do to-day, that a very large multi-million-dollar unit may be built and fail to do the job.

As a result of the widespread activity in cracking research, a number of different processes became available for licensing. Companies could choose the one best suited to their particular needs. This ability to select and combine the best features of different processes was another of the many factors that enabled the oil industry to make rapid progress. The total capacity of cracking units installed in United States refineries rose as follows:

Year	Capacity of Cracking Units in United States, barrels per day
1912	None
1920	160,000
1930	1,420,000
1940	2,360,000
1950	4,226,000

By 1930 the advent of the chemical engineer, plus the rapid growth of the industry, resulted in the development of very large refining units—notably combination cracking and crude-running units handling 40,000 to 50,000

barrels of crude per day. Perfecting these required still more research and pilot plant data, to insure optimum design and also optimum operation.

This situation gave rise to contracting firms that specialized in the construction of giant refinery units. These contracting firms not only contributed to design, but also carried out research of their own and operated pilot plants. The research became another important source of contributions to petroleum technology. As one example, The M.W. Kellogg Company contributed importantly to the development of various methods of cracking and hydroforming.

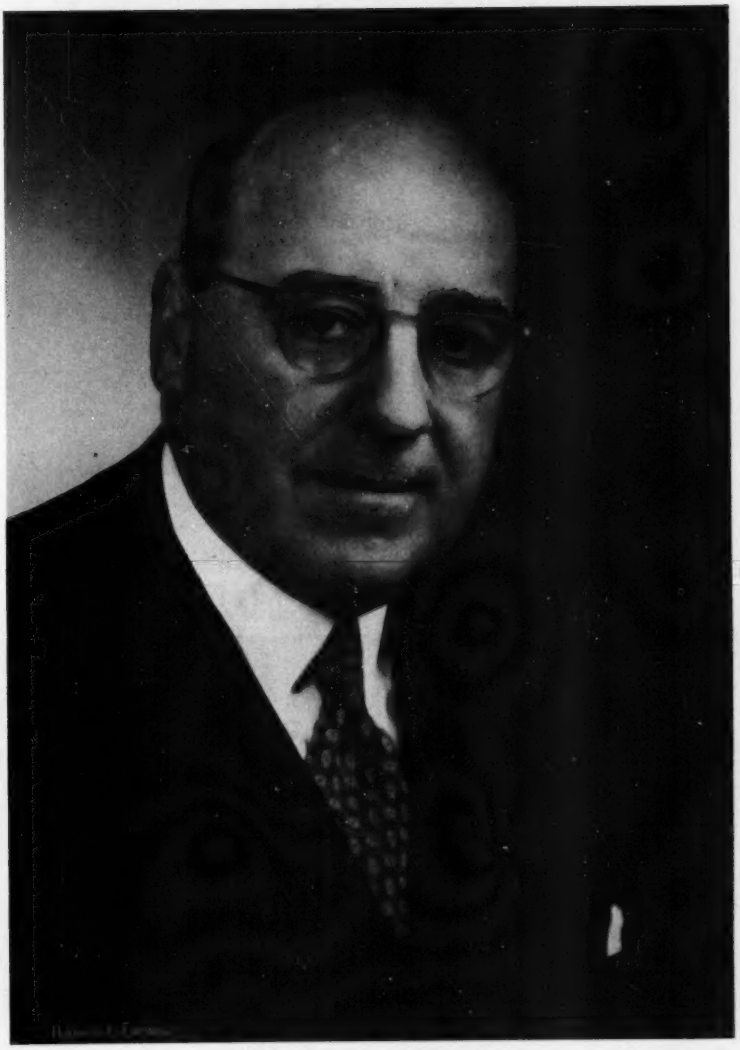
Development of Catalytic Cracking

Even twenty-five years after Burton, competitive research on cracking had two more unexpected triumphs up its sleeve. With the growing importance of catalysis in all sorts of chemical reactions, it was obvious to try to use catalysts to speed up cracking. Aluminum chloride cracking was an early development by McAfee, and rather widely used by the Gulf Oil Company, though other companies did not find it to be competitive with the thermal processes. Hundreds of solid catalysts were tried, but most of them were far too expensive for use in a field where an extra cost of a tenth of a cent per pound of product was considered high. Another basic difficulty was that anything which speeded up cracking also speeded up coke formation, and in ten or fifteen minutes the coke deposit put any good catalyst out of action. That seemed to most to constitute an insuperable obstacle. But Eugene Houdry, an independent French inventor, later assisted by the Sun Oil Company, kept plugging away until a feasible method was developed for regenerating the catalyst by burning off the coke with air in 15-minute cycles. The superior octane number of the resulting gasoline made the process very attractive.

For a time it appeared that the companies which had been collecting cracking royalties from Sun and others would soon be on the paying side in a big way. Again, however, the spirit of competition led to a frantic search for a process which would avoid the sharp temperature changes in the catalyst beds and certain other disadvantages of the Houdry process. Anglo-Iranian and Socony-Vacuum developed methods involving moving beds of catalyst. A more novel and interesting solution was fluid catalytic cracking, in which the powdered catalyst works in a fluidized bed and is transferred by air or vapour lifts between the reaction and regeneration vessels every few minutes. The coke, always one of the great problems in cracking, is converted into an asset, since most of the heat for the "fluid" process comes from burning the coke off the powdered catalyst!

Whereas the Burton process made about 1½ per cent of gasoline per hour, fluid catalytic cracking makes about 50 per cent of gasoline in the 10 or 15 seconds the gas oil vapours are in contact with the catalyst! A single modern unit moves 2,000 tons of catalyst back and forth every hour, and makes as much gasoline in a year as 500 Burton stills—but such a unit unfortunately costs more than a thousand times as much. It might be said that we owe much to our technologists, but they have made us owe more to our bankers!

Catalytic cracking was, of course, the backbone of wartime production of 100-octane aviation gasoline. To-day it is the main factor in the com-



DR ROBERT E. WILSON

CADMAN MEMORIAL LECTURER AND MEDALLIST, 1951

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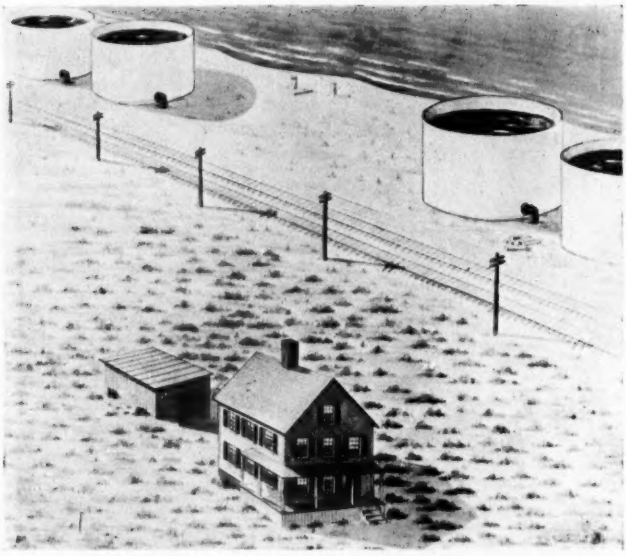
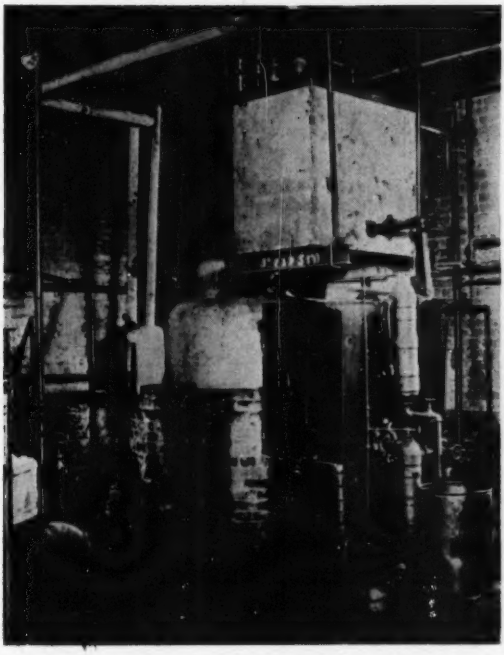
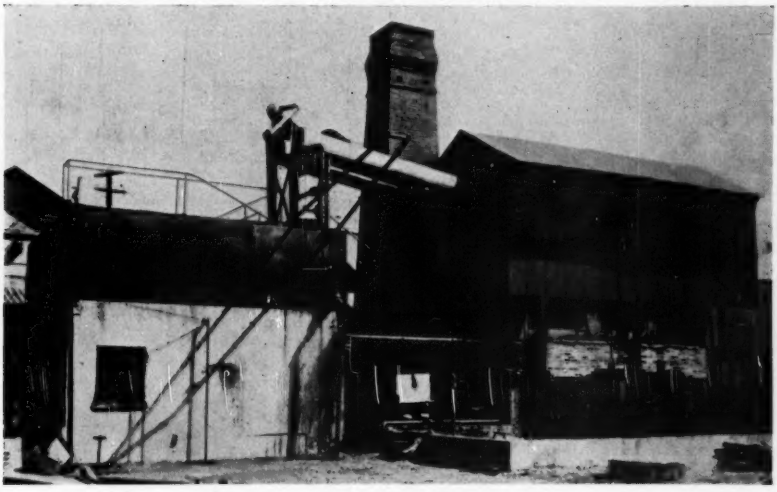


THE FARM-HOUSE USED AS FIRST OFFICE AND LABORATORY AT THE WHITING (INDIANA) REFINERY IN 1890. THE COWSHED IN THE REAR WAS USED FOR LARGER SCALE EXPERIMENTS

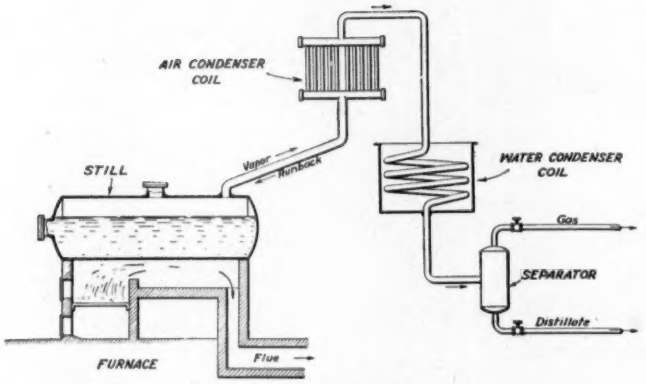
OFFICE EMPLOYEES, THE WHITING REFINERY, 1894. DR WILLIAM M. BURTON IS SIXTH FROM THE LEFT IN THE TOP ROW



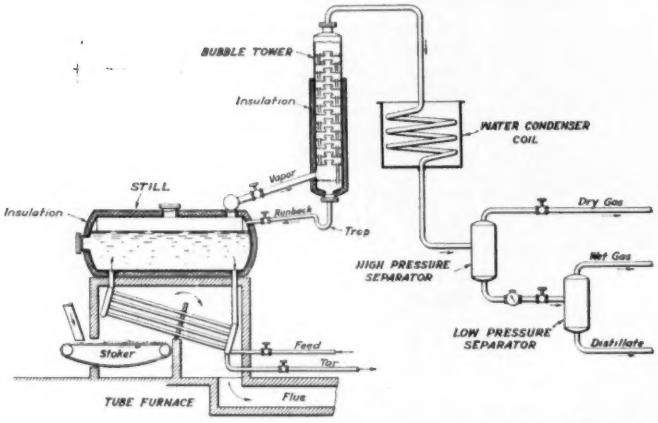
THE FIRST EXPERIMENTAL BURTON STILL, IN WHICH EXPLORATORY CRACKING WORK WAS CARRIED OUT IN 1910 AND 1911



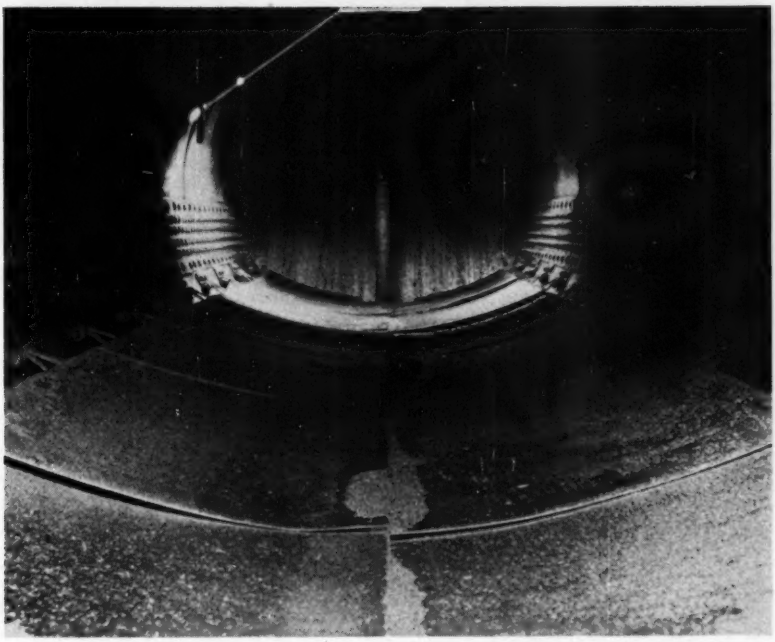
THE FIRST SEMI-COMMERCIAL BURTON STILL, USED TO ESTABLISH OPERATING CONDITIONS FOR SUBSEQUENT FULL-SCALE UNITS



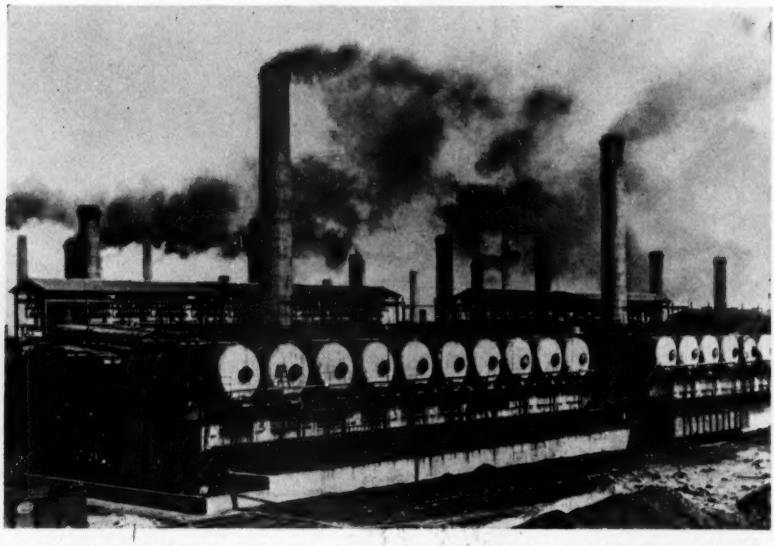
AN EARLY BURTON STILL. THIS FORM HAD A RADIATOR TO CONDENSE KEROSINE AND LIGHT GAS OIL VAPOURS AND RETURN THEM TO THE STILL FOR FURTHER CRACKING



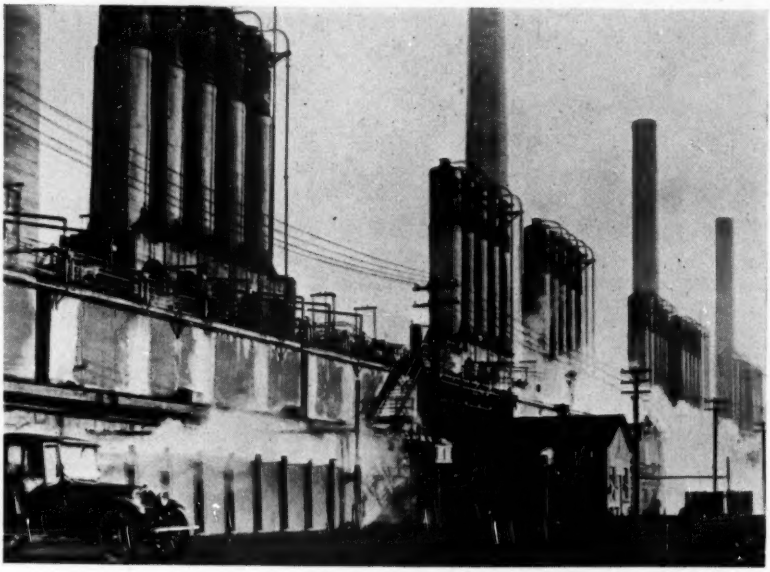
BURTON–CLARK TUBE STILL, WHICH REDUCED THE HAZARDS INVOLVED IN DIRECT FIRING OF A SHELL STILL. BUBBLE TOWERS IMPROVED THE FRACTIONATION



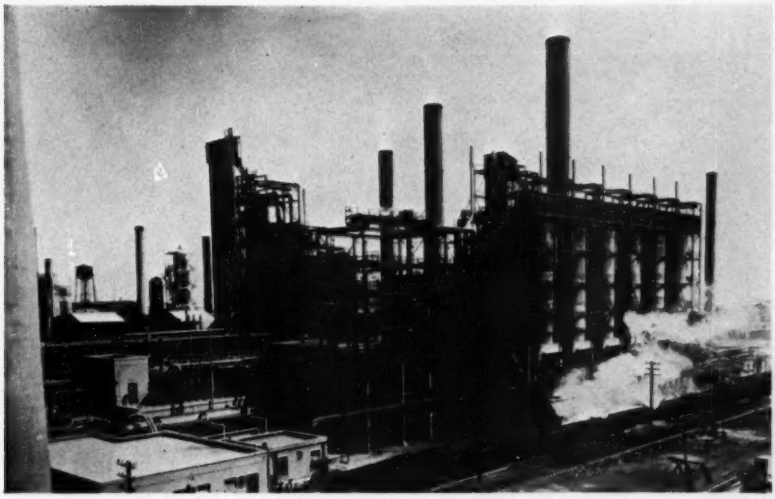
INSIDE A STILL, SHOWING THE FALSE BOTTOM PLATES DEVELOPED BY DR ROBERT E. HUMPHREYS TO MINIMIZE COKE DEPOSITION ON THE HEATING SURFACE



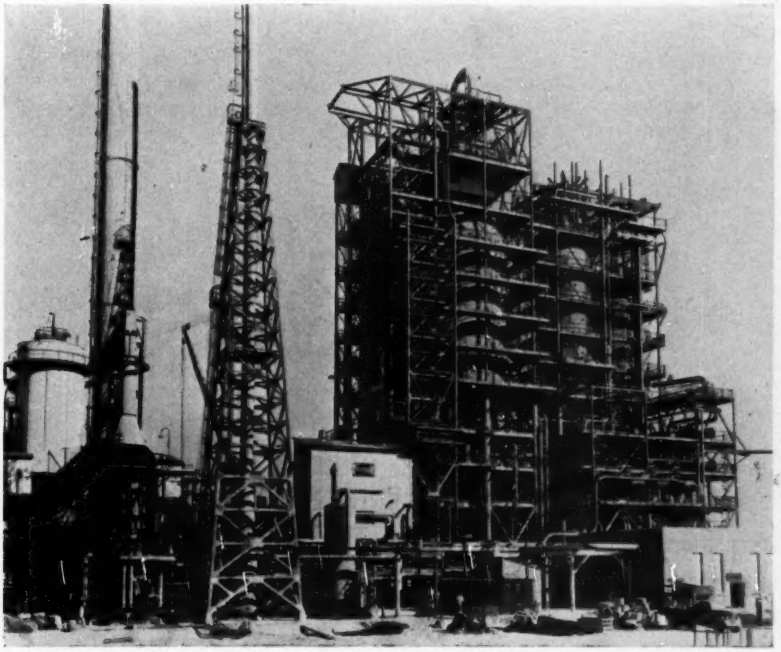
A BATTERY OF BURTON STILLS IN OPERATION. STILLS SUCH AS THESE PRODUCED MORE THAN 200 MILLION BARRELS OF GASOLINE BEFORE THEY WERE SUPPLANTED BY NEWER METHODS OF CRACKING



ROW OF BUBBLE TOWERS USED TO FRACTIONATE PRODUCTS FROM THE IMPROVED BURTON–CLARK CRACKING EQUIPMENT

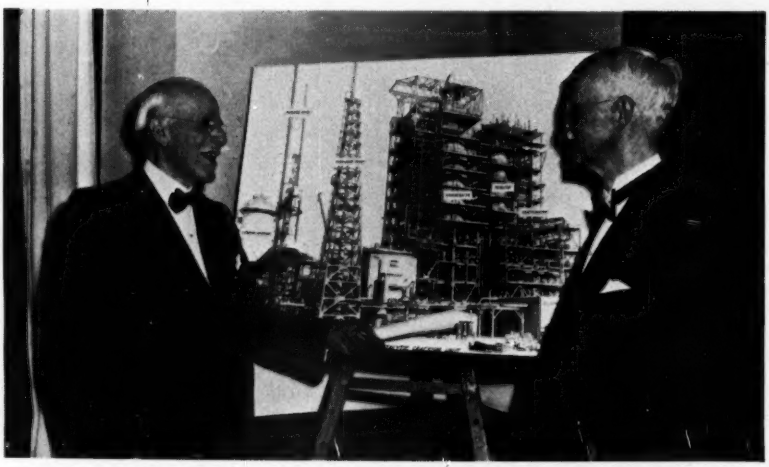


A GIANT MODERN COMBINATION THERMAL CRACKING, COKING, AND CRUDE RUNNING UNIT AT WHITING



A 30,000-B.D. FLUID CATALYTIC CRACKING UNIT AT THE WHITING REFINERY

THE WHITING RESEARCH LABORATORY OF THE STANDARD OIL COMPANY (INDIANA)



DR WILLIAM M. BURTON AND DR ROBERT E. HUMPHREYS, HONOURED AS PIONEERS IN OIL CRACKING, AT A NEWCOMEN SOCIETY MEETING, CHICAGO, OCTOBER 29, 1946

petitive octane race which is making possible automobile engines of higher compression and better efficiency.

Results Freely Available

You will note that I have been talking as if anyone could use the new advances, no matter who made them. To a large extent this was true, in spite of intense competition between different companies. The reason lies partly in the American patent system, and partly in the way the oil industry has elected to operate under that system. No discussion of competitive research would be complete without a discussion of patent and licensing policies.

The United States patent system is not radically different from the British. It grants to the inventor a monopoly for seventeen years, in return for full disclosure of his invention. There have been times, I regret to say, when our courts tended to nullify the patent laws by setting up overly severe standards of invention. At the time cracking research was getting under way, however, the courts were generally sustaining patents instead of narrowing or voiding them. There was a good chance that an inventor would acquire a valid title to his development, and so would be in a position to exercise the patent rights he had acquired by his inventiveness. These rights could be sold, traded, or leased, like any other property. The inventor, therefore, had every incentive to publish his discovery so that possible licensees—or purchasers—could learn about its merits.

Need for Cross-licensing Arrangements

Before considering the general licensing policies of the industry, it is necessary to discuss cross-licensing agreements between different owners of patent rights. These were made necessary by the vigorous research of competing firms on cracking, and the large number of overlapping patent assets which resulted. Since this subject is complicated and sometimes misinterpreted, I shall try to explain the basic principles behind it. Perhaps the safest way to do so is to use the words of the United States Supreme Court in the famous cracking patent anti-trust suit.*

"*An interchange of patent rights and a division of royalties according to the value attributed by the parties to their respective patent claims is frequently necessary if technical advancement is not to be blocked by threatened litigation. . . .*

"*A patent may be rendered quite useless, or 'blocked,' by another unexpired patent which covers a vitally related feature of the manufacturing process. Unless some agreement can be reached, the parties are hampered and exposed to litigation. And, frequently, the cost of litigation to a patentee is greater than the value of a patent for a minor improvement. . . .*

"*If the available advantages are open on reasonable terms to all manufacturers desiring to participate, such interchange may promote rather than restrain competition. . . .*"

Generally speaking, cross-licensing arrangements are not made as soon

* Standard Oil Company (Indiana) *et al v* United States of America, 283 United States Reports 163, 171 (1931).

as a new research area begins to open up. Whenever a new lead is found, the tendency is for many companies to rush in and do exploratory work. The research organizations that do the best jobs begin to make progress, and acquire patent assets.

A cross-licensing arrangement is frequently found desirable when a company learns that one or more of its patent applications is in interference with others in the Patent office. This is an indication that two or more parties have been working in a given area and have probably attained some success. If it turns out that the parties do actually have substantial patent assets, and that the claims are genuinely in conflict, a cross-licensing arrangement is usually preferable to several years of litigation and uncertainty. Also, the parties can then feel reasonably sure of having a better ultimate process by employing the best ideas of each.

Cross-licensing Stimulates Competition

For a time there was a belief in some quarters, including the United States Department of Justice, that such arrangements interfered with competition and caused a trend to monopoly. Actually, just the reverse is the case.

Cross-licensing is a great stimulant, both to progress and to competition, particularly in the technological field. Research is one method of competing. Cross-licensing increases the attractiveness of research by permitting companies to bring their new developments to actuality more rapidly and more confidently than would be the case if they faced the prospect of lengthy patent litigation.

Such cross-licensing agreements usually include the exchange of know-how as well as free or low-rate licenses. There is considerable incentive to other companies to become eligible for these advantages by doing research and so acquiring patent assets of their own. In general, companies cannot "buy their way into" cross-licensing arrangements, because they are based on a definite exchange of licenses under real patent assets. However, cash payments may be made to balance relative positions. Combinations of mere research potential in a given field are seldom made, at least among the larger companies, though a few such arrangements were entered into during the second world war at Government request, in view of the pressing need for certain synthetic products vital to the war effort, such as rubber, toluene, and high-octane aviation gasoline.

There has been sharp competition in licensing between different cross-licensing groups, as most of you doubtless know. Universal Oil Products and the Texas–Indiana group were active competitors in cracking licensing for many years. In the catalytic cracking field, Sun, Houdry, and Socony-Vacuum have been in sharp competition with another group which developed and licensed the fluid process. In the field of alkylation, Anglo-Iranian took the lead in the work on the sulphuric acid process, and Shell, Jersey, and Texas made important contributions; another group headed by Universal Oil Products, Sinclair, and Phillips, specialized in HF alkylation. In the solvent extraction of lubricating oils, different groups developed and promoted three competitive processes: chlorex, furfural, and phenol. In solvent dewaxing, propane and methyl-ethyl ketone are competing solvents.

All the important cross-licensing arrangements in the petroleum industry provide for the licensing of others, on reasonable royalty terms, by one or more of the parties to the arrangement. In addition, each party to the cross-licensing arrangement generally retains the freedom to license its own patents independently, and on such terms as it deems appropriate.

Different Licensing Policies in Various Industries

Patent licensing policies in the United States differ rather widely between industries. In some industries most companies are loath to grant any licences except at very high rates or under unusual circumstances. This tends to slow down commercial development until the patent situation is clarified, and to reduce competition in manufacture, though not in research. Some even try to rely on keeping their process secret as long as possible. At the other extreme are certain industries which, by agreement or in practice, license competing firms at little or no royalty. While some in the United States urge this as a desirable practice, from my observation it sharply reduces the amount of applied research carried out in the industry, because of the weakening of incentives—a company finds it easy to sit back and do little or nothing, because it can expect to get a free license if anything new is developed by its competitors.

The policy of the petroleum industry has been, with very few exceptions, to license any new development to all-comers at moderate, but nevertheless substantial, royalties. This policy was arrived at by a gradual process.

At the outset, Standard of Indiana realized that it would probably make more money by refusing licences and keeping its legal monopoly of making then scarce gasoline from then plentiful gas oil. However, such a policy was considered to be bad business as well as bad public relations. Refusal to licence would encourage litigation and the rise of competing processes. The first licences set rather high royalty rates, and included some other restrictions, but these were soon dropped as a matter of policy.

Licensing practices in the petroleum industry are also rather unique in the widespread use of what is called "definition" licensing. Under this method the petroleum companies, instead of licensing a specified list of patents for any *purpose*, generally desire or grant licences in a *broadly defined field*—such as thermal cracking—under *any* of the licensor's patents which may be found useful. Such a licence usually includes freedom to operate not only under any of the licensors *existing* patents, but also under related patents which may be acquired by the licensors for a reasonable number of years in the future. The result is that a licensee under one of these broad-definition arrangements is free to use any variation particularly suited to his operations, and any improvements which his licensors may make, without additional royalty. This feature has been of great benefit where processing methods are as varied as they are in the oil industry. Moreover, in general, licensors make available their designs and technical know-how to assist the licensee to arrive at the best process for his particular refinery situation. If a small refiner had to shop among many different individual patent owners, each interested in proving its patented features the most important, his problem would be much more difficult.

Definition licensing includes a right to far more than the essentials of

the process. It also includes a licence to use, in the defined process, many devices such as special pumps, alloys, fractionating columns, and so on. The breadth of the definition may be extended to include combination processes. For example, refiners holding licences under the continuous thermal cracking patents were licensed automatically when the large combination cracking and crude-running units were developed.

Benefits to the Small Company

The licensing and cross-licensing policies of the oil industry have been favourable to the small company. They have made the most up-to-date processes available to all, and so have helped the small refiners to make progress and stay in business in competition with the larger companies.

These policies have made it possible—and often desirable—for the smaller companies to concentrate their research on one or more parts of the petroleum field, where a given company may have acquired some special knowledge or advantage. Meanwhile the company can use, in its other operations, the patented developments of other companies at a cost lower than that of maintaining a complete research organization. The old view that a royalty is a sort of tribute has faded out. Most companies have found that they can make more money by adopting an improved process quickly and paying moderate royalties than by waiting for several years while they try to develop an alternative royalty-free operation. As a matter of fact, practically all the larger companies are to-day paying royalties in some fields and collecting them in others, depending on the respective patent situations.

As mentioned previously, a number of important processes have been perfected by small concerns. The cracking developments made by the Cross brothers in a small independant laboratory constitute an interesting example. They pioneered high-pressure cracking and demonstrated its feasibility and safety. Some of the most important developments in the field of gasoline antioxidants were made by research men at Mid-Continent, a small company, and by the National Benzole Association in England. The first feasible process of catalytic cracking was worked out by Eugene Houdry, an independent inventor.

The patent system also fostered the rise of organizations concerned with research and licensing alone. Universal Oil Products Company was originally formed to sue the users of the Burton process, on the mistaken assumptions that condensation under pressure was its essential feature, and that this feature was first disclosed in the old Dubbs patent on emulsion breaking. Research to prove Universal Oil Products's theories led to further research to develop a continuous cracking process. The company was eventually responsible for many important inventions in cracking and other petroleum-refining fields. It has taken in many millions of royalties, most of which has been ploughed back into research on new refining processes which could also be licensed. It is now an outstanding research and technical service organization, particularly for small refiners. To-day the company stock is in a trust, of which the American Chemical Society is the sole beneficiary. The dividends are to be used to carry out basic research in the field of petroleum.

Conditions Needed for Productive Research

To summarize, I believe that in the light of our experience, if competitive research is to give the best overall results, a number of conditions are necessary. Among these are :—

First, there must be a reasonable number of strong companies, able to attack problems aggressively from different viewpoints.

Second, there must be a sound patent system, to furnish incentives and at the same time to encourage prompt publication of research results. Competitive research kept secret for any long period is wasteful. It fails to provide opportunity for the cross-fertilization of ideas and for combining features from various sources to make the best available process.

Third, there must be freedom and willingness to license others at reasonable rates, and to enter into cross-licensing arrangements where needed. This may well include the exchange of ideas and co-operation on pilot-plant and process design work, in advance of building expensive commercial units.

Fourth, there must be no agreements on prices or in restraint of competition. The American cross-licensing arrangement is in no sense a cartel. It does not set prices, establish quotas, or divide up marketing areas. It fosters competition instead of discouraging it. While a cross-licensing arrangement might be used as a means of enforcing a cartel type of operation, this has not been done by the American oil industry—to do so would violate both our competitive tradition and our anti-trust laws.

Co-operative Research in Petroleum

The American petroleum industry believes in competitive research as the best method for most purposes. As it has evolved I believe it has avoided both the danger of too much duplication and that of too little competitive effort by different organizations with different viewpoints.

There are, however, fields in which co-operative research has been found preferable. I have already referred to the fine work of the Co-ordinating Research Council, sponsored jointly by the automobile and petroleum industries. It has carried out its research projects in various ways; by financing research at the National Bureau of Standards; through co-operative study of fuel performance in test fleets; and by assignment of various projects to the laboratories of individual companies. Projects have ranged from a determination of the desirable vapour pressure, octane number, and endpoint of motor gasoline in different cars, to a study of the effect of sulphur in gasoline on engine wear.

Research Sponsored by the American Petroleum Institute

Another body of outstanding co-operative research has been that carried out under the sponsorship of the American Petroleum Institute. The present programme really stemmed from the pioneer efforts of Van

H. Manning, director of the United States Bureau of Mines during the first world war. Shortly after the close of that war, Dr Manning accepted the position of director of research of the newly formed American Petroleum Institute and endeavoured to develop a broad programme of industry-financed research. One of his first acts was to assemble an advisory committee of scientists and engineers, to formulate problems and projects he hoped the American Petroleum Institute would sponsor. That committee included Dr Burton, Kettering, Midgeley, and a young MIT professor named Wilson!

The committee suggested a long list of problems, many of which had immediate practical importance. Dr Manning outlined a rather ambitious programme for the support of petroleum research in universities and other institutions.

However, when the proposal was submitted to the API directors late in 1920, strong opposition developed. Much of this stemmed from the fact that Dr Manning's proposals did not distinguish between fundamental problems, well adapted to co-operative research, and practical problems, the solution of which would be expected to have immediate commercial and competitive value. Many of the strong individualists on the board felt, with good reason, that such research should continue on a competitive rather than a co-operative basis. So far as I can learn, no consideration was given at that time to the possibility of selecting for co-operative attack only those fundamental long-range and expensive problems which would probably not be undertaken by competitive industry laboratories. In any case, the whole programme was turned down.

Many of the industry leaders still felt that the Institute should sponsor and support fundamental research; but the programme might never have recovered from the initial setback, had not Mr Rockefeller in 1925, and shortly thereafter the Universal Oil Products Company, made their generous propositions to contribute annually $50,000 apiece for five years to support basic petroleum research, providing the Institute would furnish the sponsorship and technical guidance. As a result, the API Board established a top-level Research Committee under the chairmanship of Walter Teagle.

Petroleum at the time had the dubious distinction of being one of the most important but least understood raw materials in the world. Hundreds of different products were made from it, but very little was known about its composition, or its method of formation and accumulation. Although many in the industry felt that more knowledge on these points would be valuable, and most of them were at least curious about these important questions, it was realized that to get the answers would be a very long and expensive job. Certainly, no single company could hope to do so. The problems were of a type which demanded a major co-operative effort extending over a period of years.

During the first five years a total of forty-one different fundamental-research projects were financed at about twenty-five different institutions throughout the United States—each project being guided by an API technical committee. Much basic information of real value was obtained. For the first time, we began to understand what we were working with. Many of the participating institutions were stimulated to do additional research in these interesting new fields.

A crisis arose when the first outside gifts ended at a very unfortunate time—in the depression year of 1931. By this time, however, the value and the propriety of co-operative research on fundamental problems were better appreciated. The Institute agreed to continue the programme on a somewhat reduced scale, raising funds on a voluntary basis from the company members of the Institute. For the next five more-or-less depression years, the expenditures averaged only about one-third of the original $100,000-a-year budget. It was also decided to concentrate on a much smaller number of more important projects, and to conduct them at institutions and under project directors who had shown particular ability to get results. The budget for this work has been steadily increased in recent years, and stands at well over $500,000 for the coming year.

Results of American Petroleum Institute Research

Results from this co-operative research on fundamental problems have been most satisfactory. Petroleum technologists throughout the world are familiar with the important work on the composition and properties of petroleum. Also, studies on the synthesis and properties of light hydrocarbons proved unexpectedly valuable during the second world war, when we needed to know how specific hydrocarbons would perform in aviation engines and how they could be made. The work on the physical and thermodynamic properties of hydrocarbons has been a great aid to the design of refinery equipment. And study of the behaviour of oil–gas mixtures at high pressures proved of enormous help in understanding the strange phenomenon of retrograde condensation in the deeper underground reservoirs where pressures are very high. Without this knowledge hundreds of millions of barrels of distillate would have been irretrievably lost in the ground. The API research has repaid the industry—and so the public —many times what it cost.

The $500,000 annual budget of the API is, so far as I can ascertain, the largest amount being spent on really fundamental research by any trade association, though some other groups do spend more on applied research. However, it is still only half of 1 per cent of what the industry is spending on competitive research.

Government Research

The question of the proper place of government in research is a difficult one. In some fields it appears to be the best available agency. Examples are agriculture and atomic energy, where the overall public interest is large, and where no private concerns exist which could or would carry out more than a small fraction of the needed research. Government research is also appropriate for making surveys of important raw-material resources, including methods of recovery, and fundamental studies of their basic characteristics. Again, the government is an appropriate agency for much of the research on standardization. One type of work that deserves increased government support is research on public health.

Even in some of the fields related to oil products, government research has seemed desirable. While the industry believes there is enough petroleum in sight for several decades, with demand growing so rapidly and new reserves

becoming harder to find, attention should certainly be given to the possibilities of synthetic liquid fuels from coal or oil shale. Pilot-plant studies in this field are quite expensive, costing more than most individual companies could reasonably be expected to undertake, especially since any resulting patents would probably expire before the processes would attain commercial importance. In spite of these attacks the industry has done considerable work in this field, and has encouraged and co-operated with the U.S. Bureau of Mines, whose recent budget for such activities has been several million dollars a year.

Two industry advisory committees have been set up to keep in general touch with this work, and samples of various crude products have been made available to industry laboratories for trying out different methods of refining, etc. The Bureau's work on the large-scale mining of oil shale with modern machinery has given much better figures for mining costs than were available previously. The Bureau has also made progress in methods of retorting oil shale and in methods of hydrogenating different kinds of coal under different conditions.

The industry generally believes that government research and pilot-plant work along these lines should be continued. However, it does not share the view of those in the Bureau who assert, with some political support, that the government should shortly proceed to build or subsidize large-scale " demonstration plants " at very high costs. Such plants would be obsolete before they were finished, could not compete without a big government subsidy, and would contribute less to the development of the processes than 5 per cent as much money spent on well-planned bench-scale and pilot-plant research and development.

Government work of this type is much more likely to be effective and of practical value if carried out in close contact with industry advisory committees of technical men.

Some Problems to Be Solved

One problem faced by industry generally is the need for much more basic research. In the United States we have in the past depended, to a greater extent than we should, on fundamental research done in Europe. We are aware of the fact that this store of basic information is being used up and must be replenished if progress is to continue. The government has recently assumed part of the obligation to encourage such research, through the formation of a National Science Foundation. The stated objective is to concentrate on fundamental science, and to aid in the training of more and better scientists. Industry is watching with interest to see whether political pressure groups will keep their hands off this programme, and whether Congress will be able to resist the demand from some quarters that the Science Foundation be diverted to solving practical problems of interest to some particular group. There is also the need to educate taxpayers as to the large public interest in basic research.

If the National Science Foundation does shift its attention to the field of applied research, it will tend to inhibit rather than encourage applied research, because private companies will generally be unwilling to enter a given field in competition with the government.

Industry Encouragement of Basic Research

Industry in the United States is already taking several steps to encourage basic research. The most direct is the carrying out of basic research in the companies' own laboratories. Such work is increasing, but is still not large in percentage. In companies—as in the nation—there are pressure groups with problems that just have to be solved right now. The people working on basic science tend to be diverted to the solving of these urgent practical problems.

Many forward-looking companies are now encouraging fundamental university research, partly through the granting of substantial fellowships for graduate work. A National Research Council survey several years ago indicates that more than 300 companies were providing a total of approximately 1800 fellowships, scholarships, or grants for research, and the number is much larger to-day. I would say that most, if not all, of the grants made by industry are made on the basis that the company does not influence the selection of recipients, and acquires no rights to the results of the research work done by the fellows.

Several universities have recently set up rather large-scale basic research projects, supported in large part by industry. Examples are the nuclear research institutes of the University of Chicago and similar undertakings at Massachusetts Institute of Technology and California Institute of Technology. Contributions by industry to these projects are now running well over $2 million a year. Company sponsorship of the three research institutes of the University of Chicago involves an annual contribution of $50,000, or $20,000 for any one institute. It may be of interest that of the funds received from industry by the universities mentioned, about 30 per cent now comes from the petroleum industry.

Some Threats to Research

I think enough has been said to demonstrate that we have a fair balance of different kinds of research in our industry, with possibly a need for more emphasis on fundamental science. We believe that our applied research has evolved into a satisfactory and productive pattern.

In view of the outstanding success of applied industrial research in the United States, and the great public benefits, it might be assumed that such research has clear sailing ahead. Actually, it faces a number of threats. Some people object to the whole idea of granting a temporary monopoly through patents and would emasculate our patent system. Others object to the competitive advantage which research gives, and wish to destroy any such advantage by compulsory licensing or other methods, thus removing the incentives for applied research. A few people fail to understand that technological unemployment is a myth so far as the overall economy is concerned; whenever there is unemployment they wish to declare a moratorium on research. It is noteworthy that they quickly subside in times of national danger. Another threat is that of government competition in the applied field, which of course would discourage research by industry, as I have already mentioned. High taxes not only reduce the funds available for research, but also reduce the incentive to develop and install new processes

when the possible profits would largely go to pay excess profits taxes. Still another threat is the drafting of technical men for non-technical military service (which I understand you were wise enough to avoid in Britain, even in wartime).

Need for Encouragement of Research

As we view the world, both ancient and modern, we see comparatively few areas in which scientific research has existed on any extensive scale. Evidently it is a plant that needs a favourable climate. We in America have been lax, as I have noted, in our encouragement of basic research. I hope to learn more, while I am here, about the methods by which Britain and Germany have in the past so successfully stimulated such research. There are many other interesting questions. What, for example, were the factors that gave Germany the lead in so many fields of research? Some, like the dyestuff industry, were fields in which some other country was once ahead of Germany. What are the reasons for, and results of, the increasing emphasis on co-operative research by various industries in several European countries? I am sure all of us would profit from a discussion of the factors behind research in various other parts of the world.

Applied research on a major scale is really a product of the past third of a century. It has revolutionized many industries and raised the standard of living more than any other development in world history. It is almost unbelievable that its continuance would be jeopardized by its millions of beneficiaries. But just when research is getting more difficult and expensive, we are faced with the widespread weakening of incentives and new threats to individual and business freedom. Research is to-day in real jeopardy unless you and I, who understand it, get its great story across to the general public. As Faraday well said, when asked by Mr Gladstone what good would come of a new discovery in electrical science, "*Why, sir, there is every probability that you will some day be able to tax it.*" And our public-utility friends to-day will add a fervent "and how"!

Let us not by our silence, or our preoccupation with scientific and industrial matters, permit the demagogues to bring about the killing of the goose that lays the golden eggs. Given a fair chance, research can contribute far more than it already has to lengthen and enrich human life.

Despite the clouds on the horizon, I am optimist enough to believe that, if we scientists do our part, neither the people nor the politicians will deprive research of the freedom and the continued opportunity to which its record of past achievement clearly entitles it.

PRESENTATION OF THE CADMAN MEDAL

Mr C. A. P. Southwell, M.C., president of the Institute of Petroleum, who occupied the chair, said :

Dr Wilson, on behalf of everyone here, I want to express our great appreciation of the stimulating and most interesting address to which we have just had the pleasure of listening.

The Cadman Memorial Medal, with which Dr Wilson's address is linked, was founded by friends in the Anglo-Iranian Oil Company of the late Lord Cadman. It was their wish to commemorate a great friend and a great oil man—one of the greatest exponents of the scientific approach to all problems in the petroleum industry. He had his heart and soul in the petroleum industry and the warmth of his personality brought him friends in every walk of life.

The Medal is a token of appreciation of Lord Cadman's service to the petroleum industry. It is awarded by the Institute of Petroleum to those who have distinguished themselves in the application of science to industry in the sphere of petroleum, irrespective of nationality or of membership of our institute.

The Council of the Institute of Petroleum unanimously approved the recommendation of their Awards Committee that the third Cadman Memorial Medal should be awarded to Dr Wilson for his outstanding contribution in a lifetime of service to the petroleum industry, in scientific development, research, and administration.

It is a tradition of the Royal Institution not to introduce the lecturer before his address, but now that the lecture, which has been so much appreciated, is concluded, I should like to be sure that everyone present knows of Dr Wilson's great contribution to science and to the petroleum industry.

In connexion with new chemical processes, Dr Wilson has published over eighty scientific papers, thus making the results of his work available to students and workers in the industry. He has nearly a hundred patents in his name, and it would be quite impossible for me to enumerate them on this occasion. As chairman of a Petroleum Industry War Council Committee and a petroleum adviser to his Government, Dr Wilson served the Allied cause with distinction during the last war.

He is the holder of two great honours in the chemical world, the Chemical Industry Medal and the Perkins Medal.

We see a great similarity between the work of the man whose name we commemorate and Dr Wilson. Like Lord Cadman, Dr Wilson started in the academic sphere, became an outstanding organizer, and then chairman of a great industrial concern. Like Cadman, he is "a man of science and a man of affairs."

We are very glad of this occasion to cement still further the bond of British and American friendship and co-operation.

I am greatly privileged, as a student of Professor Cadman as he was then, that during my year of office as President of the Institute of Petroleum, it has fallen to my lot to present to Dr Wilson the Cadman Memorial Medal.

In doing so I wish to express our hope and confidence that he may have many more active years in which to continue his great work.

It is now my very great privilege to present the Cadman Memorial Medal.

The Cadman Memorial Medal was then handed to Dr Robert E. Wilson amid prolonged applause.

DETERGENCY OF CARBON-BLACK SUSPENSIONS. PART III. IN TOLUENE

By F. H. Garner, O.B.E.* (Fellow) and A. R. W. Baddeley † (Student Member)

Summary

The rates of settling of carbon-black suspensions in toluene have been compared after the addition of stearic, oleic, and linoleic acids, and calcium oleate and linoleate. The acids have practically no effect on the rate of settling, and are not significantly adsorbed on the carbon. The calcium soaps are adsorbed, and the effect on the rates of settling is dependent on the amount adsorbed; the order found, in increasing effectiveness is calcium naphthenate, oleate, and linoleate.

A commercial additive was found to be better than any of the calcium soaps.

Introduction

In previous papers [1, 2] the results of investigations of the effect of the addition of calcium naphthenate on the characteristics of a suspension of carbon black in toluene were described. It was found that adsorption of calcium naphthenate took place according to the Langmuir equation and that a correlation existed between the additive concentration of the solution in which maximum adsorption was reached and that in which the dispersing effect of the additive on the suspension was greatest.

Test Methods

Sedimentation volumes, rates of sedimentation, and adsorption isotherms were determined in the same manner as previously.[1]

Two groups of additives have been examined in toluene solution, first the aliphatic acids, stearic, oleic, and linoleic, and secondly, the calcium soaps, calcium oleate, and calcium linoleate. For comparison a commercial additive, called additive A, was included with these.

Data on the materials used are given below :—

Stearic acid. Melting point 68·5° C. Literature value [3]: 68·5° C.

Oleic acid. Melting point 14° C. Literature value [3]: 14° C. Iodine value: 88. Theoretical: 89·9.

Linoleic acid. Melting point −11° to −9° C. Literature value [3]: −11° C. Iodine value: 175. Theoretical: 181.

Iodine values were determined by the Wijs method.

Calcium oleate and calcium linoleate were prepared from their respective acids of the above quality.

Carbon black. Micronex black—Soxhlet extracted with toluene before use.

Toluene. A.R. grade, dried with Na_2SO_4 before use.

* Department of Chemical Engineering, University of Birmingham.
† Monsanto Chemicals Ltd.

Results

Dispersion Index. The amount of settling of the suspension taking place during the first 3 hours has been taken as a measure of the degree of dispersion of the suspensions. In columns 3 and 4 of the following tables the volumes of clear solution above the suspension boundary after 3 hours and after 48 hours respectively have been recorded. The dispersion index has been calculated from this settling data over the first 48 hours by expressing

Table I

Concn of acid, g/100 ml	Concn millimoles per litre	Settling at 3 hr, ml	Settling at 48 hr, ml	Dispersion Index	Micromoles adsorbed
Stearic Acid					
0·13	4·5	8·5	12·5	32	2·5
0·26	9·0	9·5	12·9	26	2·5
0·51	18·0	8·9	13·8	36	2·5
Oleic Acid					
0·12	4·1	8·9	13·2	33	2·5
0·23	8·3	10·3	13·5	24	2·5
0·47	16·6	10·1	14·1	28	2·5
2·5	89·0	9·6	14·2	32	2·5
Linoleic Acid					
0·13	4·5	8·4	12·8	34	7
0·25	9·0	8·6	12·8	33	7
0·50	18·0	10·5	14·5	28	7
1·01	36·0	10·2	13·8	26	7

Table II

Concn of soap, g/100 ml	Concn, millimoles per litre	Settling at 3 hr, ml	Settling at 48 hr, ml	Dispersion index	Micromoles adsorbed
Calcium Oleate					
0·125	2·07	4·1	15·4	73·5	24·0
0·25	4·15	3·9	15·0	74	29·2
0·50	8·3	2·5	15·2	83·5	38·0
0·75	12·45	2·4	14·8	84	38·8
1·0	16·6	1·9	14·5	87	38·8
Calcium Linoleate					
0·125	2·1	3·7	14·5	74·5	32·5
0·25	4·15	2·8	14·8	81	44·3
0·50	8·3	2·4	14·6	83·5	51·3
0·75	12·4	1·9	12·9	85	54·2
1·0	16·6	1·5	13·7	89	51·7
Additive A					
0·25	2·5	4·6	13·6	66	34
0·5	5·0	2·5	13·8	82	52
1·0	10	2·9	14·7	80	60
1·5	15	1·5	13·7	89	83
2·0	20	1·6	14·1	89	83
2·5	25	1·8	13·9	87	83

as a percentage the ratio of the volume settled in the last 45 hours to the volume settled over the whole 48. Thus the more uniform the dispersion, the more uniform the rate of settling throughout 48 hours and the more the settling occurring during the last 45 hours and therefore the higher the dispersion index.

Comparisons of the dispersion index and the amount of additive adsorbed for each additive studied are given below in Tables I and II and Figs. 1 and 2.

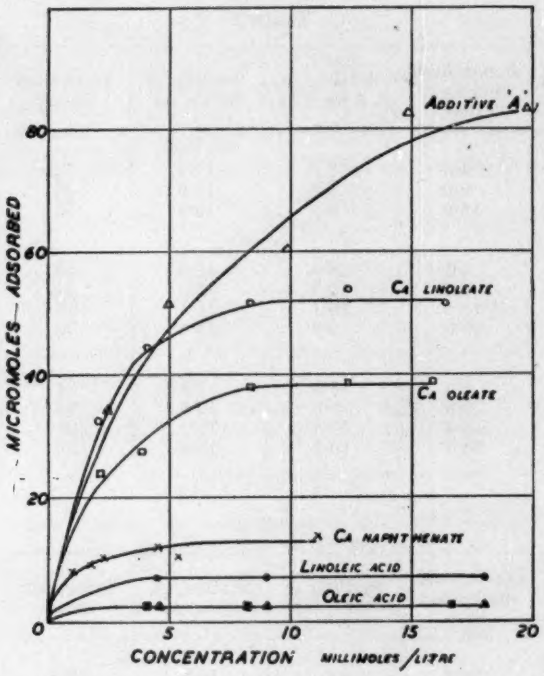


FIG. 1

ADSORPTION ISOTHERMS

With the acids it will be noted that the amount adsorbed on the carbon is very low indeed.

For calcium oleate, there is a pronounced increase in the quantity of calcium oleate absorbed until a concentration of approximately 0·01 molar is reached where a saturation effect is apparent.

For calcium linoleate, this limiting amount absorbed is also reached at a soap concentration of approximately 0·01 molar.

A comparison is given in Fig. 1 of the micromoles absorbed, plotted against concentration of additive in millimoles/litre.

Fig. 2 shows the dispersion index plotted against this same concentration.

These two graphs show that the acids have relatively no dispersion effect and that calcium naphthenate is superior to linoleic, oleic, and stearic acids.

These results are plotted from the author's previous paper.[1] However, calcium naphthenate is markedly inferior to calcium oleate, calcium linoleate, and additive A. The adsorption isotherms all reveal a saturation effect in more concentrated solutions, and adsorption follows the Langmuir equation. Too much significance cannot be attached to the absolute values for additive A, as the molecular weight is not known exactly, but the similarity between the effect of the calcium soaps and that of this commercial additive is important.

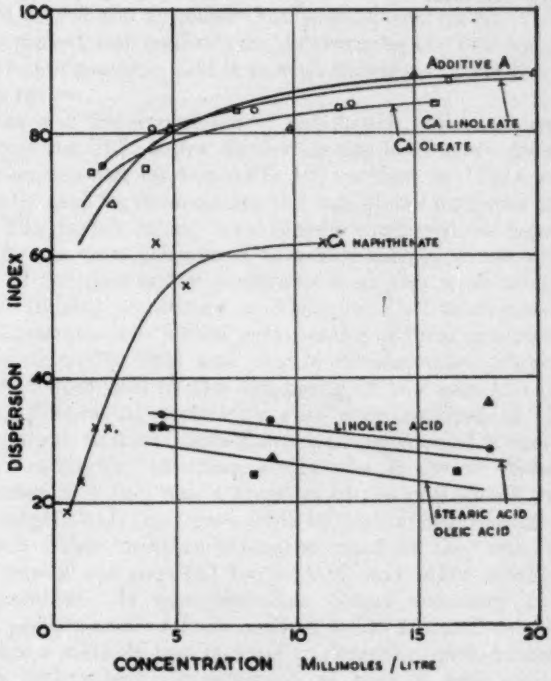


FIG. 2

DISPERSION INDEX AS A FUNCTION OF CONCENTRATION

From a comparison of Figs. 1 and 2 it may be concluded that there is a relationship between the adsorption of an additive on the particles of a suspension and the effect of the additive on the degree of dispersion of that suspension. Where an additive is strongly adsorbed, as for example calcium linoleate, then a maximum dispersion of suspended particles is observed, but if the additive is not adsorbed, *e.g.*, oleic acid, no increase in the degree of dispersion of the suspension is found. Further, it seems probable that the concentration of the solution in which a saturation value of adsorption is reached coincides with the concentration at which a maximum dispersion is obtained.

Acknowledgment

The authors wish to express their thanks to Mr F. Mohtadi for help in determining the adsorption isotherms recorded in Fig. 1.

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[2] Garner, F. H., Nutt, C. W., and Mohtadi, F. *J. Inst. Petrol.*, 1950, **36**, 292.
[3] *Handbook of Chemistry and Physics.* 31st Edition. Cleveland, Ohio: Chem. Rubber Pub. Co., 1949.

RECENT DEVELOPMENTS IN TEST METHODS FOR THE KNOCK RATING OF AVIATION GASOLINE

By JOHN OGLE *

INTRODUCTION

THE purpose of this paper is to review briefly the developments which have taken place during the last ten years in the engine testing of fuels. During this period the emphasis has been placed on the development of new and improved test methods for determining the anti-knock characteristics of aviation gasoline, and it is with this aspect that it is proposed to deal in this review.

Before the war the expression of anti-knock value was quite straightforward, since the CFR Motor method or the U.S. Army Aviation method (the latter superseded by the AFD 1-C method in 1941) were the only tests normally used for determining the anti-knock performance of aviation gasoline. The knock rating was simply expressed in terms of octane number. It was then considered that this rating, which was essentially a measure of the anti-knock performance at the weak-mixture strength used under cruising conditions in flight, was all that was required for assessing the detonation-limited performance of fuels in aero engines.

With the intensive fuel and engine development which took place immediately before and at the beginning of the war, the limitations of the existing fuel-rating technique were soon realized in the U.K. It became apparent that the anti-knock performance of a fuel varied with the mixture strength; in other words, the Motor or Aviation method octane number of a fuel was a measure of the anti-knock value at weak mixture strengths only, and gave little indication of potential performance at the much richer mixture strengths used in the aero engine when maximum power was required for take-off and other conditions of rich-mixture operation. It was, therefore, found necessary to specify the anti-knock performance of fuels both at weak- and rich-mixture strengths and to evolve a suitable test method to determine rich-mixture ratings of fuels. The satisfactory development of such a test method has undoubtedly been the most important advance in fuel-rating technique during the last decade.

DEVELOPMENT OF RICH-MIXTURE RATING TEST METHODS

(a) *Full-scale Single-cylinder Units*

The initial investigations to determine the variation of knock-limited power with mixture strength, that is mixture response, were carried out on various types of full-scale single-cylinder engines, this being considered the only way in which the anti-knock performance of a fuel in the main engine could be assessed with samples of reasonable size and without the possible complications introduced by the effect of other physical properties in multi-cylinder engines.

* Ministry of Supply—Chairman of the I.P. Knock Rating of Aviation Fuel Panel.

The single-cylinder units selected for this investigation comprised air-cooled poppet-valve (Bristol Pegasus), liquid-cooled poppet-valve (Rolls-Royce Kestrel) and, later, air-cooled sleeve-valve (Bristol Hercules) types. The operating conditions for each unit were selected to represent as closely as possible those met in the respective aircraft engines operating at take-off (rich-mixture) conditions.

Mixture response curves (see Fig. 1) relating power output in terms of b.m.e.p., as limited by incipient detonation, and specific fuel consumption, were determined on a large number of fuels of various types. The performance of each fuel was compared with that of a reference fuel known as

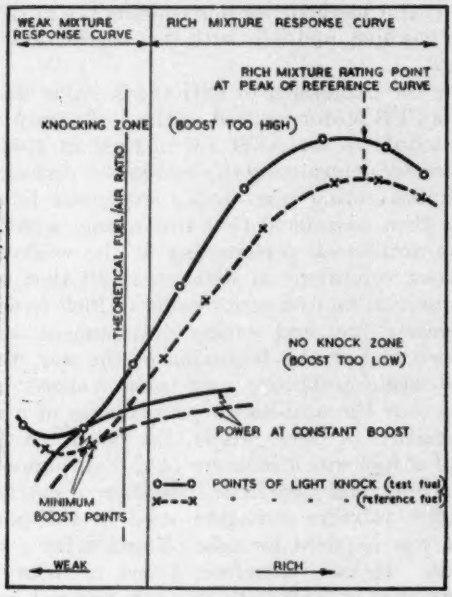
WEAK MIXTURE RESPONSE CURVE
RICH MIXTURE RESPONSE CURVE
RICH MIXTURE RATING POINT AT PEAK OF REFERENCE CURVE
KNOCKING ZONE (BOOST TOO HIGH)
THEORETICAL FUEL/AIR RATIO
MEAN EFFECTIVE PRESSURE - lbs/sq inch
NO KNOCK ZONE (BOOST TOO LOW)
POWER AT CONSTANT BOOST
POINTS OF LIGHT KNOCK (test fuel)
(reference fuel)
MINIMUM BOOST POINTS
WEAK
RICH

FIG. 1

SPECIFIC FUEL CONSUMPTION (U.K. PRACTICE) OR FUEL–AIR RATIO (U.S. PRACTICE). THE COMPLETE MIXTURE RESPONSE CURVE.

British Air Ministry 100 Octane Reference Fuel, or more briefly as BAM 100. This reference fuel consisted of a segregated shipment of 100 octane base fuel, to which was added 4·0 ml T.E.L/I.G., and was therefore representative of current fuel supplies at that time (1940).

The rich-mixture rating of a fuel was defined as the ratio, expressed as a percentage, of the b.m.e.p. of the test and reference fuel as measured at the specific fuel consumption at which the peak of the mixture response curve of the reference fuel occurred, *e.g.*,

$$\text{Rich-mixture rating} = \text{b.m.e.p. of test fuel at S.F.C. of } \frac{\text{Reference fuel peak}}{\text{b.m.e.p. of reference fuel at peak}} \times 100$$

It was clearly shown by this investigation that :—

(i) the Motor or Aviation octane number of a fuel bore little relation to its performance as measured under rich-mixture conditions on full-scale single-cylinder units;

(ii) remarkably close correlation existed between rich-mixture ratings of fuels determined on full-scale single-cylinder engines of different types.

In order to ensure adequate performance of our fuel supplies at the beginning of the war under both weak- and rich-mixture operating conditions, a rich-mixture rating requirement on Pegasus or Hercules single-cylinder units was included as a separate clause in U.K. fuel supply contracts. A rich-mixture rating clause was introduced in the U.K. specification for 100 octane fuel in 1942, requiring the rating to be not less than that of BAM 100 control fuel when tested on the single-cylinder Hercules unit running at the specified test conditions shown in Table I.

(b) *Small-scale Laboratory Test Units*

The full-scale single-cylinder knock-rating test described above necessitated the use of very costly equipment. A large fan and suitable ducting were required to deliver cooling air to the cylinder, a large compressor had to be used to provide considerable quantities of air at high pressure for boosting the engine, and a water brake of considerable size was necessary for absorbing the high power developed. A further disadvantage was that a minimum of 10 gallons of the fuel to be tested was required. For these reasons the full-scale single-cylinder test was quite unsuited as a normal inspection test.

Development of CFR Supercharge Method in U.K. Work was, therefore, started on the development of a test method using a small-scale laboratory engine to give correlation with the full-scale Pegasus and Hercules rich-mixture knock ratings. Obviously it was advantageous to choose a well-established test engine of widespread availability, and for this reason the ASTM–CFR engine was selected. The test method was similar to that used for the full-scale units in that a fixed compression ratio and variable boost pressure were used, and the rating was obtained by carrying out mixture response curves on the fuel under test and the BAM 100 reference fuel. The rating was similarly expressed in terms of knock-limited power output. A great deal of co-operative work was done in determining the engine-operating conditions required to give correlation of fuel ratings with those obtained on Pegasus and Hercules units. With the incorporation of several modifications to the unit, operating conditions were found which gave ratings agreeing fairly well with Pegasus ratings on a wide variety of fuels.

Oxygen Addition Method. Coincident with the development of the CFR supercharge method, an investigation was made on a test known as " The Oxygen Addition Method " using a modified CFR engine fitted with a means for measuring fuel flow. Oxygen was fed into the induction system of the engine, and for a given fuel setting the oxygen flow was increased until incipient knocking was obtained. An " oxygen-response curve "

was obtained by increasing the fuel flow in successive stages and determining the oxygen flow for the point of incipient knock at each stage.

Development of CFR Supercharge Method in U.S. In 1940 an invitation to co-operate in the British Supercharged CFR Correlation Programme was extended to various laboratories in the U.S., who were interested in the supply of 100 octane gasoline to the U.K. market. All expressed their willingness to take part, and samples of the Air Ministry correlation fuels were therefore sent over to the U.S. Full information on the progress of the British correlation work was at the same time made available to the U.S. laboratories.

The initial American tests were carried out on a supercharged CFR engine fitted with the Army $2\frac{5}{8}$-inch-bore cylinder, then in use for the Aviation weak-mixture test, and using a sodium-cooled cast-iron piston. The $2\frac{5}{8}$-inch-bore cylinder was chosen as it was felt that the standard under-structure of the engine was not strong enough to withstand the m.e.p.s involved when using the larger $3\frac{1}{4}$-inch-bore cylinder when supercharged. However, at the time this decision was made, very much higher supercharging pressures were contemplated than were subsequently found to be necessary.

Later an intensive programme of work was started under the direction of the CRF–AFD Committee, culminating in the introduction of the AFD 3-C Supercharge method in 1941. This method reverted to the use of the standard $3\frac{1}{4}$-inch bore CFR cylinder and was run at the test conditions shown in Table I. The test was originally intended as a measure of both weak- and rich-mixture rating. A complete mixture-response curve was obtained on the test fuel and on a sufficient number of reference-fuel blends to permit bracketing of the test fuel at its minimum boost point and at the fuel–air ratio specified for its rich-mixture knock rating. The knock rating was expressed in terms of the matching reference fuel blend, *iso*-octane in admixture with *n*-heptane or ml T.E.L/U.S.G. in *iso*-octane. It was proposed that the weak-mixture rating be determined at the minimum boost point of the test fuel occurring at a fuel–air ratio of approximately 0·06 : 1 and that the rich-mixture rating be determined at a point 50 per cent richer, *i.e.*, at a fuel–air ratio of approximately 0·09 : 1.

In practice, the determination of the minimum boost point was found to be difficult, since not only was the mixture response very rapid in this region but also small changes in mixture strength produced relatively large variations in power regardless of detonation. The definition of the fuel–air ratio at which the rich-mixture rating was determined in terms of the fuel–air ratio for minimum boost point was also unsatisfactory, since in the case of a fuel with a steep mixture-response curve a small error in the fixing of the minimum boost point had a disproportionately larger influence on the rich-mixture rating.

For these reasons it was decided that the 3-C method should be used only for the evaluation of rich-mixture performance, the rating being determined at the fuel–air ratio at which the lower bracketing reference fuel peaked. A rich-mixture rating requirement by the 3-C test method was first introduced in the U.S. Army–Navy gasoline specification in 1942, and the method has remained in use with only minor modifications until the

TABLE I

Comparison of Engine Conditions for Fuel Rating Test Methods

Method of test	Rich-mixture test methods		Weak-mixture test methods			
	Hercules	CRC–F4 (AFD 3–C)	Motor (CRC–F2)	25° Motor	U.S. Army	CRC-F3 (AFD 1–C)
Engine	Hercules S/C	Modified CFR	CFR	CFR	Modified CFR	Modified CFR
Capacity (cu. in.)	168·6	37·3	37·3	37·3	24·4	37·3
Speed, r.p.m.	2400	1800	900	900	1200	1200
Compression ratio	7·5 : 1	7·0 : 1	Variable	Variable	Variable	Variable
Spark advance, ° B.T.D.C.	16	45	19–26	25	30	35
Coolant	Air	Ethylene glycol–water	Varied with C.R. Water	Water	Ethylene glycol–water	Ethylene glycol
Coolant temp, ° F	500 *	375	212	212	330	374
Inlet air temp, ° F	230	225	75–125	75–125	Atmospheric	125
Mixture temp, ° F	—	—	300	300	No heat	220
Mixture strength	Varied with boost pressure	Varied with boost pressure	Adjusted for max knock	Adjusted for max knock	Adjusted for max thermal plug temp	Adjusted for max thermal plug temp
Knock measurement by	Aural or knock meter	Aural or knock meter	Bouncing pin	Bouncing pin	Thermal plug	Thermal plug
Induction system	Supercharged	Supercharged	Normally aspirated	Normally aspirated	Normally aspirated	Normally aspirated
Valve type : Inlet	Sleeve	—	Shrouded	Shrouded	—	—
Exhaust	Sleeve	Sodium cooled	—	—	Sodium cooled	—

* Cylinder head temperature measured by thermocouple in standard position.

present day, although the designation has been changed from AFD 3-C to CRC F.4.

Adoption of F.4 Method in U.K. With the official adoption of the F.4 method in the U.S. it was considered expedient in the interests of standardization with the U.S. to stop all development work on the British CFR Supercharge and the Oxygen Addition methods and to concentrate on the standardization of the F.4 method. Several modifications were made in the light of the experience gained during the development of the Pegasus and Hercules test methods. Chief among these were :—

(i) The use of a single reference fuel was retained. In order to overcome the weathering difficulties associated with the use of the commercial type BAM 100 control fuel and in order to line up with the U.S. system, *iso*-octane + 1·28 ml T.E.L/U.S.G., having a rich-mixture rating equal to BAM 100 control fuel, was adopted as the reference fuel.

(ii) Specific fuel consumption was plotted on the abscissa of the mixture-response curve instead of the fuel–air ratio basis specified by the U.S. It was considered that the measurement of air flow introduced an additional source of error, and was unwarranted. The measurement of i.m.e.p. adopted in the U.S. was, however, used for the U.K. method in place of b.m.e.p. in order to line up U.K. ratings with those of the U.S.

(iii) The anti-knock value was expressed as a performance rating relative to the reference fuel (*iso*-octane + 1·28 ml T.E.L/U.S.G.) instead of in terms of the matching reference fuel blend as used in the U.S. The main advantage of the U.K. method was that the rating gave a direct indication of the relative performance of fuels in a main aero-engine. (The drawback to the expression of a rating in terms of the matching reference fuel was overcome in the U.S. by the introduction of the Performance Number scale which will be discussed later.)

The F.4 method, as modified above, was included in the U.K. specifications for aviation gasoline in 1943, and has remained in use to this day.

(c) *Relationship between U.K. and U.S. Rich-mixture Ratings*

The mean peak i.m.e.p.s of blends of the U.S. reference fuels, *iso*-octane/*n*-heptane and *iso*-octane + ml T.E.L/U.S.G., have been determined on a large number of F.4 units, the values being shown in Table II, cols (i) and (ii). From these values the U.K. Rich-mixture Rating of any of the U.S. reference fuel blends can be calculated, Table II, col (iv), and a general relationship can, therefore, be established.

(d) *The U.S. Army–Navy (A–N) Performance Number Scale*

As already discussed rich-mixture fuel ratings in the U.S.A. are expressed in terms of the matching reference fuel blend. A rating expressed in this way gives no indication of the relative performance of a fuel in an engine.

To overcome this drawback an arbitrary scale, known as the Performance Number scale, has been introduced in the U.S.A. for designating the anti-

knock performance of a gasoline. This scale is based on the average performance of a variety of gasolines in both full-scale and F.4 engines related to the performance of *iso*-octane, which is assigned a performance number, of 100.

The Performance Number scale is related to the rating in terms of *iso*-octane/*n*-heptane or *iso*-octane + ml T.E.L/U.S.G., by a standard conversion table (Table II, cols (ii) and (iii)) and hence can also be related to

TABLE II

Abbreviated Conversion Tables for F4 Rating Scales

Ref i.m.e.p. p.s.i.	% *iso*-octane in *n*-heptane	A–N performance No.	% *iso*-octane + 1·28 ml T.E.L/U.S.G.
(i)	(ii)	(iii)	(iv)
136·8	93·0	80·0	67·5
145·5	95·0	84·8	71·9
161·2	98·5	94·9	79·6
168·0	100·0	100·0	83·0
	ml T.E.L/U.S.G. in *iso*-octane		
174·2	0·2	107·4	86·0
179·9	0·4	113·3	88·8
185·1	0·6	118·1	91·4
190·4	0·8	122·2	94·0
195·5	1·0	125·7	96·5
200·6	1·2	128·8	99·1
202·5	1·28	130·0	100·0
205·0	1·4	131·6	101·2
209·8	1·6	134·1	103·6
214·0	1·8	136·3	105·7
218·0	2·0	138·4	107·7
233·0	3·0	146·6	115·1
243·9	4·0	152·5	120·4
251·9	5·0	157·2	124·4
258·5	6·0	161·0	127·7

the U.K. rich-mixture rating. The scale is, therefore, merely a code for converting fuel-knock ratings in terms of reference fuels into an index which is an approximate indication of engine performance. It is, therefore, analogous to the U.K. rich-mixture rating, except for the reference point. (The performance number is the performance relative to *iso*-octane; the U.K. rich-mixture rating is the performance relative to *iso*-octane + 1·28 ml T.E.L/U.S.G., *i.e.*, a fuel having a performance number of 130.)

For convenience the Performance Number scale, although based solely on the rich-mixture performance of fuels relative to *iso*-octane, is also applied to the expression of weak-mixture ratings, the same conversion being used. Thus a fuel having an F.3 weak-mixture rating of, say, *iso*-octane + 0·5 ml T.E.L/U.S.G. and a F.4 rich-mixture rating of *iso*-octane + 3·0 ml T.E.L/U.S.G. is said to have a performance number of 115·8 at weak mixtures and 146·6 at rich mixtures. Although ratings below 100 O.N. can be expressed in terms of performance number, this is rarely done.

Development of Weak-mixture Rating Test Methods

(a) *Specification Test Methods*

CFR Motor Method. In the U.K. the CFR Motor method, also known as the CRC-F.2 method, has been, and still is, the main test used in evaluating the weak-mixture rating of aviation fuels up to and including 100 O.N.

17° *Motor Method.* The standard Motor method test possessed several disadvantages, when used for rating high-performance fuels. These were :—

(i) The high compression ratios required for rating high-performance fuels created unstable conditions of engine operation, with the result that slight fluctuations in ignition timing had a marked effect on knock intensity.

(ii) Erratic operation of the bouncing pin resulted from the higher combustion pressures.

(iii) The reference-fuel scale of *iso*-octane/*n*-heptane and *iso*-octane and ml T.E.L. did not permit the relative knock rating of high-performance fuels to be expressed numerically.

This led in 1941 to the development in the U.K. of a modified Motor method test, known as the 17° Motor method, for rating high-performance fuels. A fixed ignition setting of 17° early was adopted to allow the use of lower compression ratios to overcome unstable knocking, the bouncing-pin lower leaf spring tension was increased, and the reference fuel scale was provided by *iso*-octane/*n*-heptane + 4 ml T.E.L/I.G. calibrated in terms of *iso*-octane and *n*-heptane up to 100 O.N. and by extrapolation up to 120 O.N.

25° *Motor Method.* In 1943 the ASTM approved the following alterations to the Motor method to improve the accuracy of the test :—

(i) removal of the carburettor throttle plate;

(ii) use of Guide Tables for the micrometer setting (relating compression ratio to octane number) for standard knock intensity.

These modifications, which were also adopted in this country, rendered the 17° Motor method impractical, since without the throttle plate there was a break in the continuity of test conditions at 100 O.N. The Guide Tables could not be extrapolated to cover ratings above 100 O.N., because excessively high compression ratios and knock intensity would result.

To overcome this difficulty the ignition setting for maximum knock was determined with the throttle plate omitted and found to be 25° early. This value was adopted, and the method was accordingly known as the 25° Motor method. This method is currently specified in the U.K. for the weak-mixture rating of aviation gasolines of over 100 O.N.

U.S. Army Method. In America the bouncing pin, as used in the Motor method test, was not considered suitable for measuring the knock characteristics of aviation fuels. A method was therefore developed by the U.S. Army using a $2\frac{5}{8}$-inch-bore cylinder and incorporating a thermal plug located in the cylinder-head instead of the bouncing pin. The method was

therefore based on the temperature rise rather than on the rate-of-pressure rise associated with detonation. The operating conditions for this method are shown in Table I.

1-*C* (*F*.3) *Aviation Method.* It was obviously advantageous to use standard CFR parts as far as possible in any specification engine test, and for this reason further development in America took place along the same lines as had previously been used. This led to the introduction in 1940 of a weak-mixture test method similar to the U.S. Army method, in that the use of the thermal plug was retained, but using the standard CFR 3¼-inch-bore cylinder. This method, known as the AFD 1-C and later as the CRC-F.3 method, is still the standard specification test in America for the weak-mixture rating of aviation gasoline. Operating conditions are shown in Table I.

As previously explained in the section dealing with the U.S. Army–Navy Performance Number scale the ratings which are determined in terms of ml T.E.L. in *iso*-octane above 100 O.N. are expressed for specification purposes as Performance Numbers.

(b) *Development of Alternative Weak-mixture Rating Test Methods*

During the war years a very considerable amount of co-operative work was carried out on the development of alternative weak-mixture test methods. It was felt, particularly by the oil industry, that the existing specification test methods were excessively severe, and thereby derated certain types of fuels, particularly aromatics, to an unnecessary and uneconomical extent. The aircraft-engine maufacturers, on the other hand, considered that the existing test methods gave a reliable indication of weak-mixture performance under severe emergency conditions on existing engines or under normal operating conditions of increased severity which might be used on future engine types.

The main difficulty in the evaluation of any new test method which might be developed was the total lack of data on the relative weak-mixture ratings of fuels in the full-scale aero-engine in flight. There was therefore no direct basis upon which to correlate a new test method.

In the U.K. a project was put in hand to carry out weak-mixture knock ratings on a variety of fuel types in a fully instrumented Bristol Hercules engine in a Halifax aircraft. As, however, this would be a long-term programme, it was decided to develop a weak-mixture test on the Hercules single-cylinder unit so as to provide basic data for a laboratory test and at the same time to study the effect of engine variables on weak-mixture fuel ratings so that it would be known what alterations to test-method conditions were required, should it be necessary to obtain better correlation with the main engine flight test results when they became available.

The single-cylinder Hercules test was run under conditions equivalent to the most severe encountered in the full-scale engine when operating at Maximum Economy Cruise mixture strength. Mixture-response curves were carried out over a range of fuel–air ratios from 0·055 : 1 to 0·08 : 1 on both test and reference fuel (see Fig. 1). Ratings were expressed in terms of the ratio of the respective minimum boost pressures. A co-operative test programme was carried out by four laboratories on a wide

range of fuel types, but the reproducibility of the test was found to be very unsatisfactory, due to differences in the engine installations. A lengthy programme of work was, therefore, put in hand to obtain complete standardization of induction and exhaust systems, cylinder barrel, and head cooling system, etc. It was, however, not possible to achieve the reproducibility attained by the 25° Motor and F.3 method tests.

At the same time much effort was being put into the development of alternative laboratory scale weak-mixture test methods. Among those investigated were a test, similar in form to the Hercules test, using the F.4 test unit, an F.3 test using the Oxygen Addition method, a modified Research method, and a Variable Compression Ratio F.3 method. For the latter method an F.3 engine was fitted with a means for measuring fuel flow. Curves were plotted relating fuel flow and compression ratio for incipient detonation for the test and reference fuels. The minimum compression ratios for incipient detonation could be established from the peaks of the curves and the fuel rating expressed as the ratio of the respective minimum compression ratios for the test and reference fuels. The F.4 Weak Mixture method and the F.3 Variable Compression Ratio method were the most promising of the tests examined.

A change in outlook on the whole question of alternative weak-mixture test methods took place, however, on the completion of the Halifax flight tests referred to previously. The results of these tests showed that both the F.3 and 25° Motor method tests gave satisfactory correlation with the main engine in flight, the latter method being superior in that it was more sensitive and discriminating than the former. It was concluded, therefore, that the existing CFR specification test methods were adequate for the quality control and the experimental investigation of current aviation fuels and probably the fuels envisaged for the remaining life of the piston engine. All further development work in the U.K. on alternative weak-mixture knock-rating test methods was, therefore, abandoned.

As previously mentioned, the U.S. had originally intended that the F.4 test should serve both as a weak- and rich-mixture knock-rating test, but it had not proved possible to obtain the desired accuracy and ease of operation under weak-mixture conditions. After several unsuccessful revisions had been attempted, it was decided in America to retain the F.3 test method for the remainder of the war. It was appreciated, however, that the F.3 test began to lose significance at performance numbers above 140 and could not distinguish between fuels above 155 performance number, even though the performance of such fuels differed considerably in full-scale engines.

At the end of the war, however, there was a renewed demand in America for an improved weak-mixture test for rating high-performance fuels. A development programme was therefore put in hand by the CRC with the objective of providing a test method using only one set of equipment which would measure both the weak-mixture knock rating of fuels in the range between 100 O.N. and 160 weak-mixture performance number and also the rich-mixture rating. It was also desired to include in the test some direct determination of fuel sensitivity. A test known as the F.21 method using a modified F.4 engine has been developed to meet the above requirements, and is now being tried out in America.

Reference Fuel Scales

The picture would not be complete without some mention of the large amount of work which has been carried out both in the U.K. and America on the provision of a new reference fuel scale. The need for this has arisen from the limitations of the original octane number scale laid down in 1927 at a time when the use of fuels having an octane number superior to *iso*-octane was not envisaged.

With the introduction into service of fuels having weak- and rich-mixture ratings better than *iso*-octane it became necessary to select reference fuels of much higher knock rating. The reference fuels at present in use may be summarized as follows :—

(i) Iso-*octane*/n-*heptane* (the Octane scale), used for rating fuels up to 100 O.N. by the Motor method, F.3 method, and in the U.S.A. only by the F.4 method.

(ii) *Ml T.E.L. per U.S. gallon in* iso-*octane* (the Lead scale) used for rating fuels over 100 O.N. by the F.3 method and in the U.S.A. only by the F.4 method.

(iii) Iso-*octane*/n-*heptane* + 4 *ml T.E.L/I.G.* for rating fuels above 100 O.N. by the 25° Motor method.

(iv) Iso-*octane* + 1·28 *ml T.E.L/U.S.G.* for rating fuels by the F.4 method in the U.K.

The disadvantages of these scales are :—

(*a*) The number of different reference fuels in use lead to complexity in the expression of knock rating.

(*b*) The changeover from one set of reference fuels to another at the 100-octane level does not permit a continuous scale.

(*c*) Neither the octane nor the lead scale provides any indication of the possible performance of a fuel in an aero-engine. To overcome this drawback it has been necessary to provide the Performance Number scale.

(*d*) The lead scale is limited at its upper end to a Performance Number of about 160. This is because the incremental effect of T.E.L. on performance number decreases as the T.E.L. content is increased. For instance, the difference between *iso*-octane and *iso*-octane + 1·0 ml T.E.L/U.S.G. is 26 performance numbers, whereas the difference between *iso*-octane + 5·0 ml T.E.L/U.S.G. and *iso*-octane + 6·0 ml T.E.L/U.S.G. is only 4 performance numbers.

(*e*) Service fuels containing a constant amount of T.E.L. (4·8 to 5·5 ml/I.G.) are rated against unleaded reference fuels in the case of the octane scale or against reference fuels with variable T.E.L. content in the case of the lead scale. This is undesirable in that the T.E.L. effects the sensitivity of the fuel to which it is added to changes in operating conditions.

The requirements of a new reference fuel scale are that it should provide a continuous scale covering the complete range of knock-limited performance, likely to be encountered with service fuels, that the reference fuels should be equally applicable to weak- and rich-mixture rating, and

that the sensitivities of the reference fuels should, as far as possible, be equivalent to service fuels.

A great amount of work was carried out both in the U.K. and America on the calibration of a reference-fuel scale provided by *iso*-octane + 10 per cent toluene + 4·6 ml T.E.L/U.S.G. and *n*-heptane + 10 per cent toluene + 4·6 ml T.E.L/U.S.G. by Motor, F.3, and F.4 methods. Although the U.K. wished to adopt this scale, as it was found to meet the requirements enumerated above, international agreement to its use could not be reached, as the U.S. were anxious to complete their evaluation of the F.21 combined method for weak- and rich-mixture rating before recommending any change in the reference-fuel scales. Work has also been carried out in the U.S. on the possible use of a leaded triptane–heptane scale.

Conclusion

The progress made during the last decade in the development of new and improved engine test methods for the knock rating of aviation gasoline has been due in no small measure to the co-operation which has existed between the U.K. and the U.S. Differences in the test methods of the two countries, as for instance the adoption of the 25° Motor method in the U.K. and the F.3 method in the U.S., have to a large extent been due to the non-availability of suitable equipment in the U.K. and have not resulted in preventing interchange of supplies.

The need for new knock-rating test methods has undoubtedly been diminished by the advent of the gas-turbine engine burning fuel for which there is no anti-knock requirement. It seems unlikely that future developments in fuel-rating technique will be on a scale commensurate with the progress achieved during the last ten years; the emphasis will be rather on the improvement of existing methods.

Acknowledgment

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THE SEPARATION OF SULPHUR COMPOUNDS BY MEANS OF MERCURIC ACETATE

By S. F. Birch and D. T. McAllan *

An investigation of the chemical and physical properties of sulphur compounds likely to occur in petroleum products has, for some time, formed a subject for research in these laboratories. In the course of this, not only has a range of sulphur compounds been synthesized but some of the actual sulphur-containing bodies present in Iranian petroleum distillates have been isolated and characterized.

The highly sulphurous oil, obtained on dilution of the acid sludge resulting from the sulphuric acid treatment of Iranian gasoline and kerosine fractions, was found to consist mainly of neutral sulphur compounds and aromatic hydrocarbons.[1] Thierry,[2] who originally examined the oil, recovered from the gasoline acid sludge, demonstrated the presence of both aliphatic and cyclic sulphides identifying some of them. Recently Haresnape, Fidler, and Lowry[3] working with an oil similar to that of Thierry confirmed his findings, isolating and identifying a number of aliphatic and cyclic sulphides as well as establishing the presence of disulphides. The examination of the sulphur bodies from the corresponding kerosine acid sludge, now in progress has, however, proved more difficult. The present indications are that the compounds present consist mainly of alkyl-substituted cyclic sulphides and that small amounts of bicyclic sulphides are also present. The separation of these compounds by methods which proved effective with the lower-boiling fractions, *i.e.*, fractionation and selective adsorption, have, however, not been so successful owing to the increased number of isomers possible and other methods of separation have been sought to implement them. Of these, fractional extraction with mercuric acetate solution has proved useful. This method of separation was successfully used by one of the authors in the early investigation of the sulphur compounds from the kerosine acid sludge already mentioned, and by means of it thia*cyclo*pentane was isolated. At that time, however, owing to the relatively inefficient fractionation equipment available, little progress could be made with the higher-boiling fractions, and further investigation was postponed. In 1926 Challenger, Haslam, Bramhall, and Walkden [4] described the use of aqueous mercuric acetate for the separation of thiophens and cyclic sulphides from shale-oil fractions.

It is well known that organic sulphides readily form complexes with mercuric chloride which are normally well-defined solids, with definite melting points and chemical compositions, from which the sulphides can be recovered by refluxing with appropriate reagents, *e.g.*, dilute hydrochloric acid. These mercurichlorides are usually obtained by treatment of the sulphides with mercuric chloride in alcoholic solution when the mercurichloride separates out on cooling. By working with limited quantities of mercuric chloride, it is possible to achieve some separation of a mixture of

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organic sulphides, but the process is cumbersome. The use of aqueous mercuric acetate solution, which had previously shown promise, was accordingly further investigated. Preliminary tests were made on suitable synthetic mixtures, and the results obtained were sufficiently promising to warrant more detailed investigation. As a result, it has been found not only that aqueous mercuric acetate enables the separation of certain types of sulphides to be readily effected but also that it provides a convenient method for separating various types of sulphur compounds, *e.g.*, thiophens from sulphides, etc.

The present paper gives a general account of the preliminary results obtained in using mercuric acetate, but is by no means an exhaustive study into the possibilities of the method.

Discussion

Unlike the thioether mercurichlorides, the corresponding mercuriacetates are soluble in water. Thus when a solution of a (lower) dialkyl sulphide in petroleum ether is shaken with aqueous mercuric acetate, the sulphide passes into the aqueous layer, where it is presumably present as the mercuriacetate. Addition of a suitable water-soluble chloride such as sodium chloride immediately results in the precipitation of the insoluble mercurichloride, which can be filtered off and either purified further by crystallization or decomposed into the original dialkyl sulphide by boiling with dilute hydrochloric acid. By employing an excess of mercuric acetate over the stoichiometric quantities required to form the complex, it is possible to remove completely lower dialkyl sulphides from a hydrocarbon solution containing them. As no insoluble precipitate is formed, there is a clean separation between the aqueous mercuric acetate phase and the hydrocarbon layer.

Further experiments showed that the mercuriacetates of some sulphides, *e.g.*, the cyclic, are more readily formed than others. Therefore fractional extraction of a mixture of sulphides becomes possible by employing only sufficient of the acetate solution to combine with part of the sulphides present. Here the solubility of the mercuriacetate offers a practical advantage over the mercurichloride, since, on thorough contacting, equilibrium is set up between the mercuriacetate formed and the unchanged sulphides. It was found that a number of consecutive extractions of a mixture of sulphides with limited quantities of the acetate gave a series of extracts from which the addition of sodium chloride precipitated insoluble mercurichlorides ranging in composition from one extract to the other.* The sulphides regenerated from the extracts, either before or after the addition of sodium chloride, by heating with dilute hydrochloric acid varied in refractive index according to the separation achieved.

After the preliminary tests, more detailed experimental work was carried out to investigate the general scope of the method. The following points were examined :—

(1) efficiency of sulphur compound recovery;
(2) separation of a mixture of a dialkyl and cyclic sulphide;

* In practice it was found that unless the mercurichlorides were comparatively pure they tended to separate as a pasty solid or even liquid.

(3) separation of cyclic sulphides;
(4) separation of dialkyl sulphides;
(5) effect of molecular weight on separation of sulphides;
(6) effect of the presence of polysulphides;
(7) effect of the presence of thiophens;
(8) effect of the presence of mercaptans.

In the investigation, highly purified sulphur compounds were chosen partly because of their availability but mainly because of their refractive-index values, which enabled the efficiency of separation to be determined without difficulty from refractive-index measurements. A brief outline of the results obtained follows. It must be emphasized that all extractions were carried out at room temperature and that no experimental work was carried out on the effects of temperature variation.

Sulphur Compound Recovery

Total sulphur compound recovery was good, losses incurred being mainly due to loss of material through handling small quantities. It was demonstrated that if, instead of isolating the intermediate mercurichloride, the aqueous mercuric acetate was directly refluxed with dilute hydrochloric acid, then losses were largely eliminated. Normally, however, the intermediate mercurichloride was isolated, since the physical state of the complex (solid or partly liquid) gave an additional indication of separation efficiency. It was subsequently shown that 95 per cent of the sulphur content of a raw and recovered SO_2-extract of a light petroleum distillate could be removed by extraction with aqueous mercuric acetate solution.

Sulphides

Good separation of a mixture of a dialkyl and cyclic sulphide was achieved, and it was found that the cyclic sulphide was preferentially extracted. In this way, thia*cyclo*hexane was successfully isolated from a fraction obtained by the distillation of sulphur compounds obtained from kerosine extract tar-oil; this work will be reported more fully elsewhere.

Cyclic sulphides generally proved to be separable from one another, but the efficiency of separation was dependent on the actual sulphides present. Thia*cyclo*pentane was effectively separated from a mixture with 3-methylthia*cyclo*pentane, but little separation was possible from thia*cyclo*hexane.

Separation of individual dialkyl sulphides from each other was not efficient, and isolation of pure components did not appear to be possible.

No sulphides containing a tertiary carbon–sulphur linkage were used in the experimental work, since previous experience had shown that such compounds were susceptible to attack by mercury salts at the tertiary carbon–sulphur bond.* To confirm the fact ethyl *tert*-butyl sulphide was dissolved in light petroleum and treated with aqueous mercuric acetate solution. The sulphide was completely extracted, but on addition of

* During the purification of ethyl *tert*-butyl sulphide *via* the mercuric chloride complex, decomposition was found to occur, yielding *tert*-butyl chloride and the mercurichloride of ethyl mercaptan :—

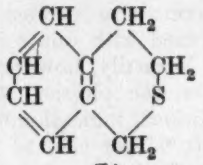
$$t\text{-}C_4H_9SC_2H_5 \xrightarrow{HgCl_2} t\text{-}C_4H_9Cl + C_2H_5S.HgCl.$$

concentrated hydrochloric acid to the clear aqueous extract the resultant product contained a large proportion of *tert*-butyl alcohol.

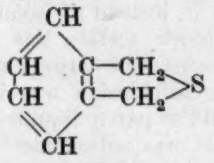
Effect of Molecular Weight on Separation of Sulphides

Two experiments were carried out without success on high-molecular-weight dialkyl sulphides which differed in the relative position of the sulphur atom in the molecule. Neither methyl *n*-decyl sulphide nor di-*n*-hexyl sulphide could be extracted from light petroleum solution by prolonged reaction with aqueous mercuric acetate solution. Addition of 25 per cent by volume of ethanol to the latter solution effected no improvement.

The reactivity of four available bicyclic sulphides with aqueous mercuric acetate solution was investigated. Isothiachroman (I) and xylylene sulphide (II) behaved similarly to the monocyclic sulphides and were completely extracted from light petroleum solution. Thionaphthen (III) was unaffected by mercuric acetate and was recovered unchanged, whilst dihydrothionaphthen (IV) only reacted slowly and to a limited extent.



(I.)



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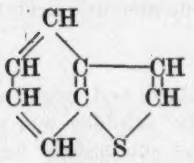
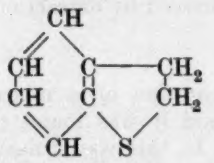


(III.)



(IV.)

On the basis of these few examples, it would appear that bicyclic sulphides react readily with mercuric acetate to form water-soluble complexes except when the sulphur atom is adjacent to a benzene ring, which seems to inhibit formation of a complex.

Polysulphides

The effect of aqueous mercuric acetate on dialkyl disulphides, trisulphides, and tetrasulphides was investigated. Extraction of a disulphide took place only very slowly, and was accompanied by decomposition which resulted in the formation of the mercuriacetate derived from the mercaptan corresponding with the disulphide. After prolonged extraction of a light petroleum solution of diethyl disulphide with a large excess of mercuric acetate solution, only 50 per cent of the disulphide had been removed, and, on regeneration of the precipitated complex from the washes, some oil was recovered which proved to be mainly ethyl mercaptan. Similar results were obtained with ethyl *iso*butyl disulphide.

Diethyl trisulphide was treated with an excess of mercuric acetate

solution and gave immediately a very small amount of white precipitate. The recovery of unchanged trisulphide from the light petroleum solution was almost quantitative, and only a trace of ethyl mercaptan was obtained from both the aqueous extract and the white precipitate.

Di-*n*-butyl tetrasulphide, in similar circumstances, was completely unaffected by aqueous mercuric acetate solution.

A mixture of dialkyl sulphide and disulphide was subsequently examined; as expected, the monosulphide was preferentially extracted leaving the disulphide practically untouched.

Thiophens

Treatment of thiophens with mercuric acetate solution generally resulted in the immediate precipitation of a highly insoluble solid complex from which the thiophen was regenerated by refluxing with dilute hydrochloric acid. Thiophen, 2-methylthiophen, and 3-methylthiophen were successfully extracted from light petroleum solution by this method, although recovery was not quite as high as with other types of sulphur compounds. 2 : 5-Dimethylthiophen, however, could not be extracted by this method, and no reaction was observed.*

A mixture of thiophen and a dialkyl sulphide (ethyl-*n*-propyl sulphide) was treated with successive washes of aqueous mercuric acetate. The sulphide was surprisingly enough removed first, and relatively little thiophen complex was precipitated until all the sulphide was removed. Since this complex is highly insoluble in water, further separation of each extract was achieved by filtration which removed the thiophen complex.

Mercaptans

No successful separation of mercaptans from the other types of sulphur compounds examined was achieved with mercuric acetate. It was therefore found necessary to remove mercaptans by other means from mixtures of sulphur compounds before attempting separation with mercuric acetate.

The product of the action of aqueous mercuric acetate with mercaptans depends on the type of mercaptan and ratio of reactants. In general, less than equimolar quantities of mercuric acetate tend to yield the corresponding mercaptide, while a large excess over the theoretical gives the mercuriacetate. Whether the product separates out or remains dissolved in one or other phase is further dependent on the molecular weight of the mercaptan, the relative concentration of the reactants, and the solvent power of the non-aqueous phase.

With excess mercuric acetate, no precipitate was obtained with ethyl mercaptan dissolved in light petroleum, but the other primary mercaptans examined yielded a solid from which the mercaptan could be recovered by heating with dilute hydrochloric acid. Removal of the mercaptan from the hydrocarbon phase was complete even with *n*-tetradecyl mercaptan and thiophenol.

* It would appear, from a survey of the literature (*vide* Steinkopf, "Die Chemie Des Thiophens," 1941), that all four hydrogens of thiophen readily react with mercuric acetate to give a solid tetraacetoxymercurithiophen. Mono-substituted thiophens are readily substituted, also under mild conditions, but 2 : 5-dimethylthiophen will only react under much stronger conditions.

Only two secondary mercaptans were examined, *e.g.*, *sec*-butyl and *cyclo*hexyl. With limited quantities of mercuric acetate, the mercaptide was preferentially formed and remained dissolved in the hydrocarbon phase; precipitation occurred only when this latter phase became saturated with mercaptide. Excess mercuric acetate favoured the formation of the mercuriacetate, and while the *sec*-butyl mercaptan derivative remained dissolved in the aqueous phase, the *cyclo*hexyl compound separated out as a white solid.

tert-Butyl mercaptan was shown to undergo decomposition involving the fission of the carbon–sulphur linkage when treated with mercuric salts.

The possibility of separating mercaptans from other types of organic sulphur compounds, by fractional extraction with mercuric acetate, thus did not appear feasible, and this was confirmed experimentally.

Experimental

Mercuric acetate solution was prepared by dissolving 319 g (1 mole) of reagent quality mercuric acetate in 1500 ml of distilled water. Any slight cloudiness was removed by the addition of a trace of acetic acid followed by filtration. The synthetic blends of sulphur compounds were dissolved in light petroleum, and all extractions were carried out at room temperature. Unless otherwise stated, all samples of regenerated sulphur compounds were washed with aqueous sodium hydroxide solution prior to drying and filtering.

Efficiency of Sulphur Compound Recovery

(*a*) Ethyl *iso*butyl sulphide (17 g), n_D^{20} 1·4450, was dissolved in 50 ml of light petroleum and shaken for 5 minutes with excess aqueous mercuric acetate solution (430 ml; 200 per cent theoretical) in three equal washes. On addition of sodium chloride, mercuric chloride complex was precipitated from the first two extracts, but the third gave only a trace of complex. The combined precipitate was separated and refluxed with 10 per cent hydrochloric acid to regenerate the sulphide (15·5 g), which was recovered in 91 per cent yield with n_D^{20} 1·4450.

This experiment was repeated as described, but the aqueous extract was directly refluxed with dilute hydrochloric acid without isolating the intermediate complex. The crude recovered sulphide was obtained in theoretical yield with n_D^{20} 1·4440.

(*b*) Thia*cyclo*pentane (17 g), n_D^{20} 1·5047, was dissolved in light petroleum and treated with successive washes of mercuric acetate solution till no further complex was precipitated on addition of sodium chloride. The sulphide (14·5 g) was recovered in 86 per cent yield with n_D^{20} 1·5042 by refluxing the complex with dilute hydrochloric acid.

(*c*) Ethyl *n*-propyl sulphide (20·5 g), n_D^{20} 1·4461 (which normally forms a stable mercurichloride of formula $R_2S.2HgCl_2$) was dissolved in light petroleum and treated with four portions of acetate solution, each equivalent to an equimolar quantity of mercury salt. On addition of sodium chloride, complex was precipitated only from the first two extracts, and from these emanated a strong smell of sulphide. The unstable mercurichloride $R_2S.HgCl_2$ had obviously been formed which tended to decompose into the

dialkyl sulphide and the stable complex $R_2S.2HgCl_2$.* The recovered sulphide (19 g), n_D^{20} 1·4460, was obtained in 92·5 per cent yield by the usual method.

(*d*) An unrefined SO_2-extract of a light petroleum distillate (2 litres), with a total sulphur content of 0·298 per cent by weight, was shaken with an excess of aqueous mercuric acetate in several batches till no precipitate was obtained on addition of hydrochloric acid to the aqueous extract. The combined aqueous layers and precipitates were refluxed with dilute acid, and a recovery was made of sulphur compounds (14·5 g) approximately equivalent to the sulphur content of the unrefined naphtha. The treated petroleum layer was washed with water, dried with potassium carbonate, and filtered. The total sulphur content, which was determined as 0·016 per cent by weight, had been reduced by 95 per cent.

Separation of a Mixture of a Dialkyl Sulphide and Cyclic Sulphide

(*a*) An equimolar mixture of ethyl *n*-propyl sulphide (16 g), n_D^{20} 1·4461, and thia*cyclo*pentane (13 g), n_D^{20} 1·5047, was dissolved in light petroleum and washed with mercuric acetate solution in batches (20 per cent of an equimolar equivalent) till no further precipitate was obtained on addition of sodium chloride. The separate precipitates were refluxed with dilute acid, and the sulphide from each was dried and filtered. Total sulphide recovery (23 g) was 80 per cent and the degree of separation achieved is illustrated by Table I.

TABLE II

Extract no.	State of precipitated complex	Weight of recovered sulphide, g	Refractive index, n_D^{20}
—	Original blend	—	1·4691
1	Crisp white solid	7·5	1·4961
2	Crisp white solid	6·0	1·4804
3	Oil, slowly solidifying	5·5	1·4519
4	Oil, quickly solidifying	3·5	1·4469
5	Yellowish solid	0·5	1·4463

(*b*) An equimolar mixture of ethyl *n*-butyl sulphide (17·5 g), n_D^{20} 1·4491, and thia*cyclo*pentane (13 g), n_D^{20} 1·5047, was dissolved in light petroleum and extracted with mercuric acetate solution as described above. Sulphur compound (25 g) was obtained in 82 per cent recovery (Table II).

* The composition and stability of the mercurichlorides of dialkyl and cyclic sulphides have been discussed previously, in two papers presented at the Spring Meeting (March 1950) of the American Chemical Society in Houston, by McAllan, Cullum, Dean, and Fidler and Whitehead, Dean, and Fidler. Usually, the stable complexes correspond to a formula $R_2S.HgCl_2$, but sometimes the stable complex has a formula $R_2S.2HgCl_2$. If, with the latter, insufficient mercuric chloride is available to form the stable complex, the unstable complex $R_2S.HgCl_2$ is initially formed which then decomposes rapidly into the sulphide and stable complex

$$2(R_2S.HgCl_2) \longrightarrow R_2S.2HgCl_2 + R_2S$$

It is, therefore, advisable when extracting sulphides with limited quantities of mercuric acetate either to add excess mercuric acetate to the aqueous extract or to regenerate the sulphide immediately with excess dilute hydrochloric acid. In this way, possible losses of sulphide from unstable complexes can be avoided.

TABLE II

Extract no.	State of precipitated complex	Weight of recovered sulphide, g	Refractive index, n_D^{20}
—	Original blend	—	1·4705
1	White solid	7·0	1·5010
2	White solid	5·5	1·4850
3	Oil, slowly solidifying	5·5	1·4522
4	Oil, slowly solidifying	5·0	1·4491
5	Yellow solid	2·0	1·4491

(*c*) A fraction, with b.p. 141·4° C and n_D^{20} 1·4930, was obtained from the sulphur compounds in kerosine extract tar-oil. A portion (15 g) was treated with successive washes of mercuric acetate solution till no precipitate was obtained on addition of sodium chloride to the aqueous extract. The various precipitates were refluxed with dilute hydrochloric acid and the regenerated sulphur compounds separately dried and filtered. Details of the separation achieved are given in Table III.

TABLE III

Extract no.	State of precipitated complex	Weight of recovered sulphur compound, g	Refractive index, n_D^{20}
—	Original material	—	1·4930
1	Crisp white solid	4·5	1·5050
2	Crisp white solid	3·5	1·5040
3	Yellow-white solid	1·5	1·4960
4	Yellow solid	0·5	1·4845

The residual light petroleum solution was evaporated down to leave a small residue (1·5 g) with n_D^{20} 1·4180. The first two extracts yielded good-quality thia*cyclo*hexane, the physical constants of which should be b.p. 141·6° C; n_D^{20} 1·5067.

Another fraction (40 g) from the same distillation boiled at 145·6° C with n_D^{20} 1·4765. On fractional extraction with mercuric acetate solution various fractions of sulphur compound were obtained as shown in Table IV.

TABLE IV

Extract no.	State of precipitated complex	Weight of recovered sulphur compound, g	Refractive index, n_D^{20}
—	Original material	—	1·4765
1	Crisp white solid	8	1·4960
2	White oily solid	8	1·4875
3	Yellow paste	6·5	1·4750
4	Orange paste	3	1·4600
5	Orange syrup		

The residual light petroleum solution had a "higher ketone" type of odour, and was treated with aqueous semicarbazide hydrochloride and

sodium acetate to yield a solid semicarbazone which was recrystallized from aqueous ethanol to a melting point of 122° to 123° C. No depression of melting point was obtained on mixing with the semicarbazone of methyl *n*-amyl ketone, and analysis confirmed that the extracted compound was indeed methyl *n*-amyl ketone.

Analysis : Found—C = 56·3; H = 10·0; N = 24·2 per cent.
Calc for $C_8H_{17}N_3O$ C = 56·11; H = 10·00; N = 24·54 per cent.

The isolation of ketones from acid sludges has been reported by several authors.[5]

Evaporation of the petroleum solvent yielded an aromatic oil (7 g), which distilled in the range 140° to 144° C with n_D^{20} 1·4660. The distillate burned with a smoky flame, decolorized bromine water slowly, and gave no test for sulphur. It was probably mainly aromatic hydrocarbon not completely removed from the tar-oil by previous acid treatment.[1]

Separation of Cyclic Sulphides

(*a*) Thia*cyclo*pentane (13 g), n_D^{20} 1·5047, and 3-methylthia*cyclo*pentane (15 g), n_D^{20} 1·4924, were treated in the usual way with successive portions of mercuric acetate solution. Recovery of sulphides (25 g) was 89 per cent (Table V).

TABLE V

Extract no.	State of precipitated complex	Weight of recovered sulphide, g	Refractive index, n_D^{20}
—	Original blend	—	1·4982
1	White solid	8·5	1·5004
2	Semi-solid oil	7·5	1·4984
3	White solid	7·0	1·4954
4	Yellowish solid	2·0	1·4926
5	White solid	0·5	1·4924

(*b*) An equimolar mixture of thia*cyclo*hexane (15 g), n_D^{20} 1·5061, and 3-methylthia*cyclo*pentane (15 g), n_D^{20} 1·4924, was dissolved in light petroleum and treated with mercuric acetate solution. An 81 per cent recovery of cyclic sulphides (27·5 g) was made (Table VI).

TABLE VI

Extract no.	State of precipitated complex	Weight of recovered sulphide, g	Refractive index, n_D^{20}
—	Original blend	—	1·4989
1	Oil, solidifying quickly	8·5	1·4998
2	Oil, solidifying quickly	7·5	1·4990
3	White solid	7·0	1·4982
4	Yellow solid	3·0	1·4959
5	White solid	Negligible	—

Separation of Dialkyl Sulphides

Equimolar quantities of di-*iso*propyl sulphide (17 g), n_D^{20} 1·4388, and ethyl *n*-butyl sulphide (17 g), n_D^{20} 1·4491, were processed in the usual way. The sulphides (27·5 g), were recovered in 81 per cent yield (Table VII).

TABLE VII

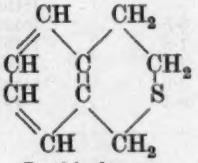
Extract no	State of precipitated complex	Weight of recovered sulphide, g	Refractive index, n_D^{20}
—	Original blend	—	1·4442
1	Yellow solid	6·5	1·4428
2	Yellow solid	6·0	1·4428
3	Yellow solid	6·5	1·4433
4	Yellow solid	4·5	1·4443
5	Semi-solid oil	4·0	1·4458

Effect of Molecular Weight on Separation of Sulphides

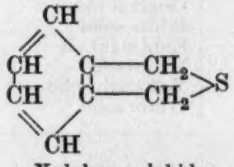
Methyl *n*-decyl sulphide and di-*n*-hexyl sulphide were each dissolved in light petroleum and repeatedly extracted with mercuric acetate solution. No complex was precipitated on addition of salt to either extract, and the sulphides were recovered unchanged from the petroleum solution by evaporation.

The experiment was repeated with a 25 per cent alcoholic solution of mercuric acetate, but again no extraction was achieved.

The effect of mercuric acetate solution on bicyclic sulphur compounds was also investigated. Only four such compounds were available, *i.e.*:

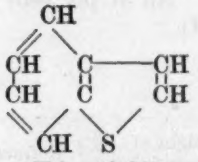


Isothiachroman

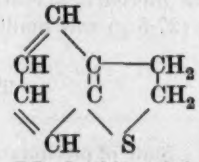


Xylylene sulphide

and



Thionaphthen



Dihydrothionaphthen

Isothiachroman and xylylene sulphide behaved similarly to the monocyclic sulphides, and the mercuriacetate was completely soluble in the aqueous phase. Addition of hydrochloric acid precipitated the mercurichloride from which the original sulphur compound was recovered in theoretical yield by refluxing with hydrochloric acid.

Unlike the parent body, thiophen, thionaphthen was unaffected by mercuric acetate and was recovered unchanged from the light petroleum solution. Dihydrothionaphthen reacted only very slowly with the extracting solution, and even after prolonged shaking with excess mercuric acetate, only 40 per cent was removed from petroleum solution. It is impossible to draw any definite conclusions from the study of only four compounds, but it would appear that when the sulphur atom is adjacent to the benzene ring no complex is formed.

Effect of the Presence of Polysulphides

(*a*) Diethyl disulphide (6 g) n_D^{20} 1·5072, was washed six times with an equimolar equivalent of mercuric acetate solution. The addition of sodium chloride to each extract precipitated a yellow powder which did not melt even on strong heating. On evaporation of the petroleum solvent from the oil layer, crude disulphide (3 g) was recovered unchanged. The yellow precipitate was continuously refluxed with dilute hydrochloric acid, and a small amount of low-boiling oil (2 g) slowly separated. This oil, which boiled mainly about 35° C, proved to be ethyl mercaptan (n_D^{20} 1·4313) with perhaps a trace of disulphide as indicated by the refractive index, n_D^{20} 1·4440.

Treatment of ethyl *iso*butyl disulphide (7·5 g), n_D^{20} 1·4951, gave a similar result. From this, it appears that a disulphide is not readily extracted by mercuric acetate solution but it slightly decomposed, and, on the subsequent addition of sodium chloride, yields the mercurichloride of the mercaptan corresponding with the disulphide

$$\text{R.S.S.R.} \xrightarrow[2\text{NaCl}]{\text{Hg(O}\overline{\text{Ac}})_2} 2\text{RS.HgCl} + 2\ \overline{\text{Ac}}\text{ONa}$$

(*b*) Diethyl trisulphide (6 g), n_D^{20} 1·5654, was washed with a large excess of mercuric acetate solution in several batches. The clear aqueous layers were filtered off, and a very small amount of white precipitate in the oil layer was separated. On evaporation of the petroleum solvent, trisulphide (5 g) was recovered having n_D^{20} 1·5550. Treatment of the aqueous extracts with dilute hydrochloric acid yielded only a trace of ethyl mercaptan, as did the white precipitate on similar treatment. Di-*n*-butyl tetrasulphide on similar treatment was completely unaffected by aqueous mercuric acetate solution.

(*c*) Ethyl *iso*butyl sulphide (12 g), n_D^{20} 1·4450, and diethyl disulphide (12 g), n_D^{20} 1·5072, were dissolved in light petroleum and washed with a 75 per cent molar equivalent of mercuric acetate solution. The individual extracts were refluxed directly with dilute hydrochloric acid and the recovered sulphur compound was not washed with alkali (Table VIII).

TABLE VIII

Extract no.	Weight of recovered sulphur compound, g	Refractive index, n_D^{20}
—	Original blend	1·4740
1	5·5	1·4470
2	5·0	1·4540
3	3·0	1·4590

The solvent was distilled from the oil layer to leave residual disulphide (10 g) with n_D^{20} 1·4950. The total recovery of sulphur compound (23·5 g) was 98 per cent.

Effect of the Presence of Thiophens

(*a*) Thiophen (7 g), n_D^{20} 1·5286, was treated with an excess of mercuric acetate solution (300 ml), and the solid complex which was immediately precipitated was filtered off and washed with light petroleum. The dried complex was refluxed with dilute hydrochloric acid and yielded recovered thiophen (5 g), n_D^{20} 1·5170, in 71·5 per cent yield. Further treatment of the light petroleum filtrate with acetate solution gave no further precipitate, and only a trace of thiophen was recovered from the aqueous filtrate.

3-Methylthiophen (10 g), n_D^{20} 1·5202, was similarly treated, and from the precipitated complex was obtained recovered material (7 g), n_D^{20} 1·5150, in 70 per cent yield. 2-Methylthiophen (10 g), n_D^{20} 1·5201, also gave a white precipitate from which the recovery (7 g), n_D^{20} 1·5180, was 70 per cent. A small amount of 2-methylthiophen (1 g), n_D^{20} 1·5140, was recovered from the aqueous extracts.

(*b*) 2 : 5-Dimethylthiophen (11 g), n_D^{20} 1·5123, was shaken with excess mercuric acetate solution, but only a trace of precipitate was formed. No sulphur compound was recovered from the aqueous extracts, but evaporation of the petroleum solvent yielded unchanged dimethylthiophen (10 g) with n_D^{20} 1·5070. (2 : 5-Dimethylthia*cyclo*pentane was completely extracted in similar circumstances.)

(*c*) An equimolar mixture of thiophen (7 g), n_D^{20} 1·5286, and ethyl *n*-propyl sulphide (8 g), n_D^{20} 1·4461, was washed with a slight excess of mercuric acetate solution in three portions. The first wash gave only a little precipitate, and the petroleum layer was treated with two further washes to give a voluminous precipitate in each case. The aqueous extracts were separately refluxed with dilute acid, as was the combined precipitate. Sulphur compound (12·5 g) was recovered in 83·3 per cent yield (Table IX).

TABLE IX

Source of sulphur compound	Weight of recovered sulphur compound, g	Refractive index, n_D^{20}
Original blend	—	1·4775
1st aqueous extract	7	1·4458
2nd aqueous extract	0·5	1·4475
3rd aqueous extract	0	—
Insoluble precipitate	5	1·5090

Effect of the Presence of Mercaptans

Mercaptans react rapidly and completely with aqueous mercuric acetate solution. Ethyl, *n*-propyl, *n*-octyl, and *n*-tetradecyl mercaptans were separately dissolved in light petroleum. After a few minutes shaking with a slightly over-theoretical excess of acetate solution, the mercaptan had completely reacted and the petroleum layer was doctor negative in every case. Thiophenol reacted in an analogous manner.

The complex formed by ethyl mercaptan was completely soluble in the aqueous phase, but with increase in molecular weight the mercuriacetate was precipitated as a white solid. Both *n*-octyl mercaptan and thiophenol were recovered quantitatively from the appropriate complex by refluxing with dilute hydrochloric acid. On similar treatment of the complex from *n*-tetradecyl mercaptan, only a trace of mercaptan distilled over, presumably due to its low volatility.

Only two secondary mercaptans were examined, *e.g.*, *sec*-butyl and *cyclo*hexyl mercaptan. With excess mercuric acetate, *sec*-butyl mercaptan yielded the mercuriacetate which remained dissolved in the aqueous phase from which a precipitate was obtained by the addition of sodium chloride. The *cyclo*hexyl mercaptan was similarly treated with excess reagent and gave an immediate precipitate of the mercuriacetate. In each case the hydrocarbon layer contained no residual mercaptan and no solid. On working with less than theoretical quantities of mercuric acetate, however, it was found that no precipitate was formed and the aqueous later contained no mercury salt. Subsequently it was demonstrated that the corresponding mercaptide had been formed which dissolved in the hydrocarbon phase from which it was precipitated only at saturation-point.

tert-Butyl mercaptan decomposed on treatment with mercuric acetate, undergoing fission at the carbon-sulphur linkage as evidenced by the formation of *tert*-butyl chloride and alcohol on heating with hydrochloric acid.

Conclusions

(1) Aqueous mercuric acetate is an effective reagent for isolation of the sulphur compounds present in the lower-boiling petroleum fractions. Certain type-separations of the organic sulphur compounds can also be achieved.

(2) Fractional extraction with saturated aqueous solutions of mercuric acetate provides an efficient method of separating dialkyl sulphides from cyclic sulphides, the latter being preferentially extracted.

(3) It is possible to separate mixtures of some cyclic sulphides, but the degree of separation is dependent on the identity of the components of the mixture.

(4) Dialkyl sulphide mixtures cannot satisfactorily be resolved by this method.

(5) Dialkyl disulphides are not readily extracted, but are slowly decomposed by the action of mercuric acetate and eventually yield mercaptans as a breakdown product. Dialkyl trisulphides and tetrasulphides are substantially unaffected by mercuric acetate.

(6) Thiophens react with mercuric acetate solution to form an insoluble precipitate, from which the thiophen can be regenerated. 2 : 5-Dimethylthiophen, however, does not react. Thiophen can thus be separated from dialkyl and cyclic sulphides.

(7) Mercaptans react rapidly and completely with mercuric acetate solution, but the product formed is dependent on the type of mercaptan and the molecular ratio of the reactants. In general, excess reagent forms the mercuriacetate, while limited amounts of reagent favour the formation of the mercaptide, which dissolves in the non-aqueous phase. Mercuric

acetate solution does not satisfactorily separate mercaptans from mixtures of organic sulphur compounds. Any mercaptans present in such mixtures must first be removed by other methods.

Acknowledgment

Our thanks are due to the chairman and directors of the Anglo-Iranian Oil Co. Ltd., for permission to publish these results.

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TESTS FOR THE CONSISTENCY OF EXPERIMENTAL ISOBARIC VAPOUR–LIQUID EQUILIBRIUM DATA

By E. F. G. Herington *

Summary

The use of isobaric vapour–liquid equilibrium data in the design of precision fractionating equipment is discussed briefly, and attention is drawn to the lack of a convenient test for the consistency of isobaric equilibrium measurements, particularly for systems where the boiling points of the components differ appreciably. A test for the consistency of binary data based on the evaluation of the area of the plot of $[(\log \alpha_{1\cdot2} - \log (p_1^\circ/p_2^\circ)]$ against x_1 from $x_1 = 0$ to $x_1 = 1$ is presented where $\alpha_{1,2}$ is the relative volatility ratio of component 1 with respect to component 2; p_1°, p_2° are the vapour pressures of pure components 1 and 2 at the boiling point of the solution containing a mol fraction x_1 of component 1. Comparison of a quantity which can be derived from the areas on this plot with a parameter which can be read from a table given in the text using the lowest boiling point (t_i° C) in the isobaric data and the total boiling range (θ) of the solutions enables a decision to be made upon the probable consistency of the data. The effect on the proposed test of the deviation of the vapours from ideal gas behaviour can be estimated. The appropriate extension of the procedure to isobaric ternary systems is indicated.

The method is applied to vapour–liquid equilibrium values for twenty-four systems recorded in the 3rd edition (1950) of the "Chemical Engineers' Handbook," by J. H. Perry, and the data on ten of these systems are shown to be of doubtful consistency; *e.g.*, see Tables III and IV.

Introduction

The advances made during recent years in the theory and practice of precision fractional distillation have necessitated the determination of accurate vapour–liquid equilibrium data. The examination of such experimental measurements for consistency before undertaking the rather laborious design calculations is now customary. Many liquid hydrocarbon mixtures obey Raoult's Law approximately, so that in the past the lack of information on individual mixtures has not greatly hampered the development of precise fractionating techniques in the petroleum industry. The production of chemicals other than hydrocarbons by petroleum refineries, however, and the adoption of separation techniques such as azeotropic and extractive distillation which depend on the non-ideality of the system now require the petroleum engineer to undertake a detailed study of vapour–liquid equilibria.

Fractional distillation is normally carried out under conditions of constant pressure, and hence isobaric equilibrium data are required for plate-to-plate calculations. The Gibbs–Duhem equation and various integrated forms of this relationship, such as the Margules, Van Laar, and Scatchard equations strictly apply only to systems under both constant temperature and pressure, but these equations may be used to investigate the results obtained in the study of systems under constant temperature alone, provided the molar volume of the liquid is negligible compared with that

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of the vapour. For a recent discussion on the nature of this approximation see Clark.[1]

The differential form of the Gibbs–Duhem equation is rarely used by chemical engineers to test the reliability of data because of the practical difficulty of measuring accurately the slopes of lines based upon experimental observations, but particular integrated solutions of this equation such as those mentioned above are frequently employed. The derivation of these special equations ensures that if isothermal data fit these equations the observations must be thermodynamically consistent (subject to the molar volume condition referred to above), but unfortunately the converse is not necessarily true. Equations of this type (*i.e.*, Margules, Van Laar, and Scatchard equations) have often been employed to examine isobaric data, and the changes in the activity coefficients with temperature have been ignored. Such a procedure is attended by some uncertainty for it is not obvious how closely isobaric experimental results should fit these expressions.

A rigorous test of the thermodynamic consistency of isobaric data is only possible if all the relevant enthalpy terms as well as equilibrium measurements are available, but because the labour in obtaining this mass of information is prohibitive such a complete test of data has rarely, if ever, been attempted. The test for the consistency of isobaric vapour–liquid equilibrium data which will be described here is of wide generality (*i.e.*, does not depend on a particular solution of the Gibbs–Duhem equation) and is well suited for examining volatility data cast in a form convenient for design calculations. The only information required additional to the vapour–liquid compositions and the vapour pressures of the pure components is the boiling range of the solutions and the lowest boiling temperature. Any analysis of experimental isobaric results which does not employ measured enthalpy values will not suffice to establish inconsistencies in the data with absolute certainty, but the suggested test will indicate if a system requires further investigation before the data can be accepted.

The tests presented are similar to those recently described for isothermal data by Herington,[2] by Coulson and Herington,[3] and by Herington.[4] The extension of this method to the examination of binary isobaric data is first described, and the necessary theoretical treatment is subsequently developed which leads to Table II, where the expected behaviour of isobaric data is summarized. A method of computing the effect of the departure of the behaviour of the vapours from the ideal gas laws is presented. Finally, the published information on a number of systems is examined.

Method of Testing Binary Isobaric Data

The relative volatility ratio ($\alpha_{1\cdot2}$) is a very convenient parameter for use in plate-to-plate design calculations. This ratio is defined by the expression $\alpha_{1\cdot2} = (y_1/x_1)(x_2/y_2)$, where y_1 and y_1 are the mol fractions of components 1 and 2 in the gas and x_1 and x_2 are the corresponding quantities for the liquid.

It is assumed initially that the vapours obey the ideal gas laws, and when this condition is satisfied identity 1 is valid, where γ_1 and γ_2

$$[\log \alpha_{1\cdot2} - \log (p_1^\circ/p_2^\circ)] \equiv \log (\gamma_1/\gamma_2) \quad . \quad . \quad . \quad (1)$$

are the activity coefficients of components 1 and 2 with respect to the pure liquids at the same temperature as standard states and with the compositions expressed as mol fractions. Identity (1) is valuable because it expresses the relationship between $\alpha_{1\cdot2}$, which is useful in design calculation, and (γ_1/γ_2) which is of considerable thermodynamic significance. The necessary modification of (1) when the vapours are non-ideal is given later.

To test the consistency of binary data the function $\log(\gamma_1/\gamma_2)$ is calculated by means of the identity 1 from the experimental measurements for a series of liquid compositions covering the range $x_1 = 0$ to $x_1 = 1$, and the results are plotted against x_1. Usually a curve of the type shown in Fig. 1

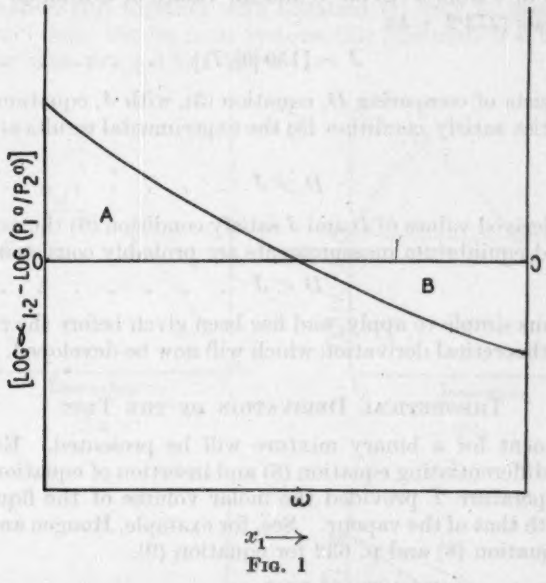

FIG. 1

will be obtained. The integal, I, given by equation (2) is next computed; due care being taken of the algebraic signs of the areas.

$$\int_0^1 [\log \alpha_{1\cdot2} - \log(p_1^\circ/p_2^\circ)]dx_1 = I \quad . \quad . \quad . \quad . \quad (2)$$

For example in Fig. 1 the integal, I, is equal to the area A minus the numerical value of the area B. The arithmetic sum of the area A and area B is also required where no notice is taken of the algebraic signs of the areas, and this sum taken as positive will be designated Σ. The percentage deviation, D, expressed by equation (3) is next computed,

$$(100\,|I|)/\Sigma = D \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

where $|I|$ is equal to the value of I taken with a positive sign.

The total boiling range (θ) in degrees Celsius of the solutions under the isobaric conditions studied is required. As an example θ equals the difference between the boiling points of the pure components under the

prevailing pressure if the system does not form an azeotrope, while it equals the difference between the boiling point of the azeotrope and the lower-boiling component if a maximum-boiling azeotrope is formed. If the system forms a minimum-boiling azeotrope, θ equals the difference between the boiling point of the higher-boiling component and that of the azeotrope. The lowest-boiling point (T_i) in ° K of the solutions in the composition range $x_1 = 0$ to 1 must also be extracted from the experimental observations. The lowest-boiling point (T_i) corresponds to the absolute boiling point of the lowest-boiling component in the system unless a minimum-boiling azeotrope is present when it corresponds to the boiling point of that mixture. The function J, given by equation (4), is computed and is tabulated in Table II for values of θ from 5° to 50° C and for values of t_i from 50° to 250° C, where T_i equals $(273{\cdot}2 + t_i)$.

$$J = [150\,|\theta|/T_i] \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (4)$$

The test consists of comparing D, equation (3), with J, equation (4), and if these quantities satisfy condition (5) the experimental results are probably inconsistent.

$$D > J \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (5)$$

while if the derived values of D and J satisfy condition (6) the experimental vapour–liquid equilibrium measurements are probably consistent.

$$D < J \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (6)$$

The test is thus simple to apply, and has been given before the rather more complicated theoretical derivation which will now be developed.

Theoretical Derivation of the Test

The treatment for a binary mixture will be presented. Equation (7) obtained by differentiating equation (8) and insertion of equation (9) is true for any temperature T provided the molar volume of the liquid is small compared with that of the vapour. See, for example, Hougen and Watson,[5] p. 649 for equation (8) and p. 632 for equation (9).

$$\begin{aligned} \ln(\gamma_1/\gamma_2) &= (1/R)[(1/T)(d\Delta G^E/dx_1)]_T \\ &= (1/R)[(1/T)(d\Delta H/dx_1) - (d\Delta S^E/dx_1)]_T \quad . \quad . \quad . \quad (7) \end{aligned}$$

$$\Delta G^E = RT \sum_{i=1}^{2} x_i \ln \gamma_i \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (8)$$

$$\sum_{i=1}^{2} x_i d(\ln \gamma_i) = 0 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (9)$$

Here ΔG^E is the excess free energy of mixing and is equal to the difference between the actual free energy change accompanying the formation of the liquid mixture containing x_1 mol fraction of component 1 from the pure liquid components and the free energy change which would accompany the formation of an ideal mixture of the same composition; ΔH and ΔS^E are the corresponding heat and entropy terms. The activity coefficients, γ_1 and γ_2, have been defined above.

As an approximation ΔH and ΔS^E will be treated as functions solely of x_1 and independent of T over the temperature range covered by the

isobaric data. Under these limitations equation (7) can be integrated to give equation (10), remembering that T is a function of x_1 for isobaric data.

$$\int_0^1 \ln(\gamma_1/\gamma_2)dx_1 = (1/R)[(\Delta H/T) - \Delta S^E]_{x_1=0}^{1} + (1/R)\int_{T_2^\circ}^{T_1^\circ}(\Delta H/T^2)dT$$

$$= (1/R)\int_{T_2^\circ}^{T_1^\circ}(\Delta H/T^2)dT \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (10)$$

In equation (10), T_2° is the boiling point in ° K of component 2 and T_1° that of component 1 for the constant pressure being considered and the integal on the right-hand side is to be taken along the composition line from $x_1 = 0$ to 1. If ΔH is known as a function of x_1 for the prevailing boiling points, equation (10) together with equation (1) may be used to test the vapour–liquid data, but for most systems this information is not available, and we may then proceed as follows.

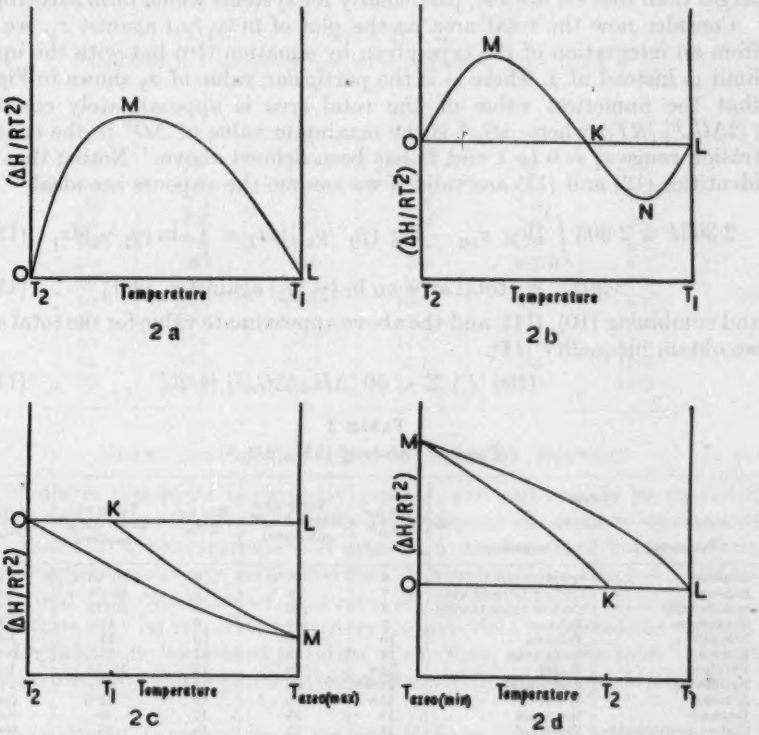

FIG. 2

Figs. 2*a*, 2*b*, 2*c*, and 2*d* illustrate some possible forms for the graph of $(\Delta H/RT^2)$ plotted against T. The value of the integal on the right-hand side of equation (10) is given in Fig. 2*a* by the area OML, in Fig. 2*b* by the area OMK minus area KNL, in Fig. 2*c* by the area OMK (*i.e.*, by area OML minus area KML) and in Fig. 2*d* by area KML. The Figs. 2*c* and 2*d*

illustrate the curves applicable to systems containing maximum and minimum boiling-point azeotropes respectively. For the present purposes we are interested only in the magnitude of the integal in equation (10) regardless of sign, and it is clear that in every example the inequality given by (11) is true.

$$(1/R)\left|\int_{T_1^\circ}^{T_1^\circ}(\Delta H/T^2)dT\right| < (1/R)\,(|\Delta H_m|/T_i^2)\,|\theta| \quad . \quad (11)$$

Here $|\Delta H_m|$ is the numerically largest value of ΔH which occurs in the concentration range $x_1 = 0$ to 1; $|\theta|$ is the difference between the lowest and highest boiling point of the isobaric data measured on the absolute scale, and T_i is the lowest boiling point expressed in degrees absolute. For example, in Figs. 2*a* and 2*b* $|\theta|$ is equal to $T_1 - T_2$; in Fig. 2*c* it equals $[T_{\text{azeo. (max)}} - T_2]$, while in Fig. 2*d* it equals $[T_1 - T_{\text{azeo. (min)}}]$. The function on the right of the inequality (11) will usually be several times larger than that on the left, particularly for systems which form azeotropes.

Consider now the total area on the plot of $\ln(\gamma_1/\gamma_2)$ against x_1, we see from an integration of the type given by equation (10) but with the upper limit ω instead of 1, where ω is the particular value of x_1 shown in Fig. 1, that the numerical value of the total area is approximately equal to $(|2\Delta G_m^E|/RT_i)$ where ΔG_m^E is the maximum value of ΔG^E in the concentration range $x_1 = 0$ to 1 and T_i has been defined above. Noting that the identities (12) and (13) are valid if we assume the vapours are ideal

$$2{\cdot}303I \equiv 2{\cdot}303\int_0^1[\log\ \alpha_{1\cdot2} - \log\ (p_1^\circ/p_2^\circ)]dx_1 \equiv \int_0^1 \ln(\gamma_1/\gamma_2)dx_1 \quad (12)$$

$$2{\cdot}303\Sigma \equiv [\text{total area on } \ln(\gamma_1/\gamma_2) \text{ against } x_1 \text{ plot}] \quad . \quad (13)$$

and combining (10), (11), and the above approximate value for the total area we obtain inequality (14).

$$(100\,|I|)/\Sigma < 50\,|\Delta H_m/\Delta G_m^E|\ |\theta/T_i| \quad . \quad . \quad . \quad (14)$$

TABLE I

Typical Values of $|\Delta H_m/\Delta G_m^E|$

System		Ref for ΔG_m^E	Temp at which ΔG_m^E measured	Ref for ΔH_m	Temp at which ΔH_m measured	$\left\|\frac{\Delta H_m}{\Delta G_m^E}\right\|$
Component 1	Component 2					
Benzene	*cyclo*Hexane	6	70° C	6	30–70° C	2·9
Benzene	Carbon tetrachloride	7	70	7	40–70	1·6
*cyclo*Hexane	Carbon tetrachloride	8	70	8	40–70	2·5
Chloroform	Ethanol	9	45	9	45	1·1
Ethanol	Toluene	10	35	10	35	1·0
Ethanol	*iso*-Octane	11	25	11	25	0·5
Pyridine	Water	12 *a*	80	13	12–13	1·5
Pyridine	Acetic acid	12 *a, b*	80	14 *b*	20	2·0
Ethanol	Water	15 *a*	50	16	42·5	0·6
Benzene	Methanol	17	35	17	35	0·9
Carbon tetrachloride	Methanol	18	35	18	35	0·6
Ethylene bromide	Chlorobenzene	19	approx 100	19	approx 100	2·7
Ethylene bromide	1-Nitropropane	19	approx 100	19	approx 100	1·2
n-Pentforane	*n*-Pentane	20 *c*	262–292° K	20 *c*	262–292° K	1·0
Ethanol	Acetone	21	40° C	22	25° C	2·5

Notes on Table I

(*a*) Calculated from vapour-liquid compositions by the equation: $\Delta G_m^E = 1{\cdot}152RT\Sigma$ (see p. 459 for definition of Σ).

(*b*) Calculated from vapour-liquid compositions (see (*a*) above) treating acetic acid as the double molecule $(CH_3{\cdot}COOH)_2$ (see p. 467).

(*c*) Re-calculated from the partial molar quantities tabulated.

The next step is to derive a maximum probable value for the ratio $|\Delta H_m/\Delta G_m^E|$ by examining actual systems for which these parameters have been measured. Table I sets out such values derived from published data.

The maximum value of $|\Delta H_m/\Delta G_m^E|$ in Table I is 2·9, and the mean value for these 15 systems is 1·5. Since these systems comprise mixtures of all types and contain polar and non-polar liquids, it appears probable that a value of 3·0 for the function $|\Delta H_m/\Delta G_m^E|$ will rarely be exceeded.

Inserting $|\Delta H_m/\Delta G_m^E|$ equal to 3 in the expression (14) yields (15), which is the basis of the proposed test.

$$(100\,|\mathrm{I}|)/\Sigma < [150\,|\theta|/\mathrm{T}_i]$$

i.e.,

$$D < J \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (15)$$

For convenience the function J is tabulated in Table II in terms of θ and of t_i, where t_i is the minimum temperature in ° C. ($T_i = 273{\cdot}2 + t_i$.)

TABLE II

Tabulation of Function $\mathrm{J} = (150\,|\theta|/\mathrm{T}_i)$ *where* $\mathrm{T}_i = (273{\cdot}2 + \mathrm{t}_i)$

θ	t_i° C				
	50	100	150	200	250
5	2·3	2·0	1·8	1·6	1·4
10	4·6	4·0	3·5	3·2	2·9
15	7·0	6·0	5·3	4·8	4·3
20	9·3	8·0	7·1	6·3	5·7
25	11·6	10·1	8·9	7·9	7·2
30	13·9	12·0	10·6	9·5	8·6
35	16·2	14·0	12·4	11·1	10·0
40	18·6	16·1	14·2	12·7	11·5
45	20·9	18·1	16·0	14·3	12·9
50	23·2	20·1	17·7	15·9	14·3

EXTENSION OF THE TEST TO TERNARY SYSTEMS

The above treatment is perfectly general, and can readily be extended to ternary systems. Tests for the consistency of ternary isothermal data based on the measurement of areas at a constant mol fraction of one component have already been described (Herington [4]), as also has a test for isothermal data at a constant mol ratio of two components (Herington [23]). These tests may be extended to ternary isobaric data, provided the percentage deviations are expressed in terms of the total areas, for then Table II may be used and the criterion of consistency (15) can be employed.

THEORETICAL INVESTIGATION OF THE EFFECT OF THE NON-IDEALITY OF THE VAPOURS

The effect of the departure of the vapours from ideal gas behaviour is usually ignored in chemical-engineering practice when attempts are made to fit experimental data to the Margules, Van Laar, or Scatchard equations. The result of deviations from ideality in the gas upon the proposed test for consistency will now be examined.

Equation (16) is the fundamental expression for these considerations where

$\bar{\alpha}_1 = (RT/P) - \bar{V}_1$; $\alpha_1 = (RT/P) - V_1$; $\bar{\alpha}_2 = (RT/P) - \bar{V}_2$; $\alpha_2 = (RT/P) - V_2$

and $\bar{V}_1$ and $\bar{V}_2$ are the partial molar volumes of component 1 and 2 in the mixed vapour, V_1 and V_2 are the molar volumes of the pure component 1 and 2; p^* is to be taken in the limit as $p^* \to 0$, and P is the total pressure.

$$\ln(\gamma_1/\gamma_2) = \ln \alpha_{1\cdot2} - \ln(p_1^\circ/p_2^\circ) - (1/RT)\left[\int_{p^*}^{P} \bar{\alpha}_1 dp - \int_{p^*}^{p_1^\circ} \alpha_1 dp + \int_{p^*}^{p_2^\circ} \alpha_2 dp - \int_{p^*}^{P} \bar{\alpha}_2 dp\right] \quad (16)$$

Two special cases are of interest.

Effect of Small Deviation from the Gas Laws

Detailed treatment of the effect of small deviations is hampered because of the lack of experimental observations on the P–V–T behaviour of mixed vapours at temperatures near the normal boiling points of the liquids concerned. A convenient measure of small deviations from ideality is afforded by the second virial coefficient, B. Thus for component 1, equation (17) will apply where B_{11} is the second virial coefficient of component 1 and V_1 is the molar volume of the vapour.

$$V_1 = (RT/P) + B_{11} \quad . \quad . \quad . \quad . \quad . \quad . \quad (17)$$

For pure component 2 there is similar expression involving V_2 and B_{22}. Equation (18) is the corresponding exact expression for the binary vapour mixture where V is the volume of vapour containing n_1 and n_2 mols of component 1 and 2, and B_{12} is a constant for given components.

$$V/(n_1 + n_2) = (RT/P) + [B_{11}y_1^2 + 2B_{12}y_1y_2 + B_{22}y_2^2] \quad . \quad (18)$$

In the almost complete absence of information on B_{12} it is common practice to assume that the relationship given by equation (19) is valid.

$$2B_{12} = B_{11} + B_{22} \quad . \quad . \quad . \quad . \quad . \quad . \quad (19)$$

Combination of equations (19) and (18) leads to equations (20) and (21).

$$V_1 = \bar{V}_1 = (RT/P) + B_{11} \quad . \quad . \quad . \quad . \quad . \quad (20)$$

$$V_2 = \bar{V}_2 = (RT/P) + B_{22} \quad . \quad . \quad . \quad . \quad . \quad (21)$$

Inserting equations (20) and (21) in equation (16) and integrating with respect to p yields equation (22)

$$\ln(\gamma_1/\gamma_2) = \ln \alpha_{1\cdot2} - \ln(p_1^\circ/p_2^\circ) + (1/RT)[B_{11}(P - p_1^\circ) - B_{22}(P - p_2^\circ)] \quad (22)$$

Integration of equation (22) with respect to x_1 for isobaric conditions yields equation (23).

$$\int_0^1 \ln(\gamma_1/\gamma_2)dx_1 = \int_0^1 [\ln \alpha_{1\cdot2} - \ln(p_1^\circ/p_2^\circ)]dx_1 + \int_0^1 (1/RT)[B_{11}(P - p_1^\circ) - B_{22}(P - p_2^\circ)]dx_1 = 2{\cdot}303[I + Z] \quad (23)$$

where $Z = (1/2{\cdot}303)\int_0^1 (1/RT)[B_{11}(P - p_1^\circ) - B_{22}(P - p_2^\circ)]dx_1$

Comparing equation (23) with equation (12) we see that the integal Z must be algebraically added to I to obtain $\int_0^1 \log(\gamma_1/\gamma_2)dx_1$ when the vapours are non-ideal, and since this latter integal should be employed in equation (15), it follows that in these circumstances the condition for consistency is given by inequality (24).

$$(100\,|I + Z|)/\Sigma < (150\,|\theta|/T_i)$$

i.e.,
$$D\text{ (corrected)} < J \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (24)$$

where
$$D\text{ (corrected)} = (100\,|I + Z|)/\Sigma$$

The small contribution of small departures from ideality to Σ has been ignored in the derivation of equation (24).

The virial coefficients, B_{11} and B_{22}, are functions of T, and T itself is a function of x_1 for isobaric data, but as we are mainly interested here in the magnitude of Z, a simplified approximate form of Z may be obtained by treating B_{11} and B_{22} as constants and by taking each equal to the value of B at the boiling points of the respective components under the pressure being considered. Similarly, T will be treated as constant and equal to T_i. Hence we may write the approximate relation for Z given by equation (25), where P is the constant total pressure and where p_1° and p_2° can be obtained from the boiling points of the various solutions together with the dependence of the vapour pressures of the pure components on temperature.

$$Z \simeq (1/2{\cdot}303RT_i)\left[(B_{11} - B_{22})P - \int_0^1 (B_{11}p_1^\circ - B_{22}p_2^\circ)\right] \quad (25)$$

From a study of actual examples using equations (24) and (25), it is shown on p. 468 that the numerical value of Z is usually small compared with that of I, so that in practice the contribution from Z to equation (24) can often be ignored.

Effect of Adopting an Erroneous Molecular Weight in the Calculations

The correct molecular weight to use in the calculation of the liquid mol fraction in the study of vapour–liquid equilibria when the ideal gas laws are assumed to apply is clearly that which corresponds to the molecular weight of the component in the vapour. In some published examples of the study of vapour–liquid equilibria a molecular weight corresponding to a simple chemical formula has been employed, although it may be known that the vapour is polymerized. For example, a molecular weight of 60 has occasionally been used for acetic acid, although it has been shown that the vapour consists mainly of dimer. If an incorrect molecular weight is used for component 1 and if otherwise the vapours obey the ideal gas laws, then $\bar{\alpha}_1$ and α_1 in equation (16) will no longer be zero and relationship (1) will no longer be valid. The magnitude of the error will depend on the extent by which the molecular weight used differs from the true value and also on the ratio of log (P/p_1°) throughout the concentration range. Calculation for particular systems shows that if a molecular weight of one-half the correct value be employed for component 1 and if log (P/p_1°) differs appreciably from zero, then the error in D arising from this cause may be appreciable.

Quick Method of Computation

Computation of the areas on the $[\log \alpha_{1\cdot2} - \log (p_1^\circ/p_2^\circ)]$ *against* x_1 *plot*

Application of the proposed test is facilitated by the use of a convenient method for estimating areas on the plots of experimental data. Any method of quadrature may be employed, but Gauss' method (see, for example, Margenau and Murphy [24]) is both simple and accurate. A useful practical device is to employ a sheet of transparent plastic $WXYZ$ ruled with full

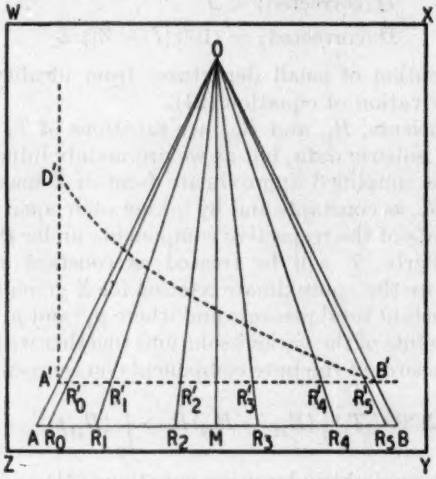

Fig. 3.

lines as shown in Fig. 3. The relative dimensions on this sheet should be as follows :—

$$AR_0 = R_5B = 0{\cdot}033765AB$$
$$AR_1 = R_4B = 0{\cdot}169395AB$$
$$AR_2 = R_3B = 0{\cdot}380690AB$$

The plastic may be ruled by eye using a steel scale to graduate the lines as accurately as possible. In a typical model $AB = 15$ cm, $OM = 14$ cm; but the transparency can be made any convenient size. Suppose it is now required to calculate the area within the dotted lines $A'D'$, $D'B'$, $B'A'$ on Fig. 3. The transparency is placed on the curve as shown with AB parallel to $A'B'$ and the ordinates at R_0', R_1', R_2', R_3', R_4', and R_5' parallel to $A'D'$ are read from the curve; let these be $y_0, y_1 \ldots y_5$. The required area can now be computed with the aid of the machine using equation (26)

$$\text{Area} = A'B'[0{\cdot}08566(y_0+y_5) + 0{\cdot}18038(y_1+y_4)+0{\cdot}23396(y_2+y_3)] \quad (26)$$

Examination of Some Published Data for Consistency

The data on binary equilibria listed on pp. 573–5 of " Perry's Handbook," 1950 edition,[25] has been tested for consistency by the method developed in this paper. All the binary vapour-liquid systems presented in that book have been examined, except for the following mixtures for which insufficient

data are given, *i.e.*, the systems acetaldehyde–water; chloroform–methanol; nitric acid–water. The data on the system benzene–ethylene dichloride was not analysed because the values published had been calculated on the assumption that the system is ideal, although in fact this system is not ideal; see Coulson, Hales, and Herington.[26] The following errors in the data as published were noted and corrected before carrying out the analyses, the listed mol fractions for the acetic acid–water system refer to water and not acetic acid as stated; the columns giving vapour and liquid compositions for the ethylene glycol-water system should be interchanged.

The required values of $\log (p_1^\circ/p_2^\circ)$ were computed by plotting $\log p_1^\circ$ and $\log p_2^\circ$ against $(1/T)$, using published figures, and by reading off from the graphs the logarithm of the pressures appropriate to each boiling point. The vapour pressures of organic liquids were taken from the collected data of Stull,[27] the vapour pressures of water from Kaye and Laby,[28] and the vapour pressures of liquid oxygen and nitrogen from Dodge and Davis.[29] For the system carbon tetrachloride–benzene at 760 mm and for the system nitrogen–oxygen at 760 mm and 3800 mm for which no boiling points are given the function $\log (p_1^\circ/p_2^\circ)$ was assumed to vary linearly with the mol fraction between the compositions $x_1 = 0$ to $x_1 = 1$. The mol fractions for the systems containing acetic acid were recalculated on the basis of a molecular weight of 120 for the acid before carrying out the computation. The results of applying the test are summarized in Table III. Some deviation from the expected behaviour will result from small experimental errors, and in the light of experience gained in the application of the test to isothermal data an uncertainty of 10 units in D may be arbitrarily assigned to this source (*e.g.*, see ref. 26) Allowance may be made for the effect of these small deviations on the test by postulating that $(D - J)$ must exceed 10 units before the data be considered as inconsistent, and in Table III such systems have been marked with an asterisk. No general relationship connecting the errors in the various $\alpha_{1\cdot2}$ values with a given uncertainty in D can be derived, but if additional assumptions are made an equation connecting these quantities can be deduced. Thus, if the plot of $[\log \alpha_{1\cdot2} - \log (p_1^\circ/p_2^\circ)]$ against x_1 is a straight line and if the $\log \alpha_{1\cdot2}$ values are all equally high or all equally low by the same amount, a small uncertainty of (ΔD) in D will results from a percentage error in $\alpha_{1\cdot2}$ of approximately $2{\cdot}30\ (\Delta D)\Sigma$. For example, in the system carbon disulphide–carbon tetrachloride (Table III) where $\Sigma = 0{\cdot}0551$, the percentage error in $\alpha_{1\cdot2}$ will be 1·3 per cent when (ΔD) equals 10 units if the above assumptions are satisfied. However, if the plot of $[\log \alpha_{1\cdot2} - \log (p_1^\circ/p_2^\circ)]$ against x_1 is not a straight line or if the $\log \alpha_{1\cdot2}$ values are not all equally in error, then the percentage errors in some of the $\alpha_{1\cdot2}$ values may be greater or less than $2{\cdot}30\ (\Delta D)\Sigma$, but nevertheless this function does serve to indicate the order of magnitude of the errors in the volatility ratios which might give rise to an uncertainty of (ΔD) in D.

For the systems containing acetic acid it is necessary to consider how closely the assumption of a molecular weight of 120 for the acid accounts for the volumetric behaviour of the vapour. That this treatment of the values for acetic acid is adequate is shown by a study of the isothermal data on the system acetic acid–pyridine (Zawadiski [12]), where a percentage deviation, D, of 12·5 per cent was found when a molecular weight of 120

was employed for the acid. In view of this observation the isobaric data on the systems acetic acid–benzene and acetic acid–water quoted in Table III are almost certainly inconsistent.

In conclusion, it should be noted that the plot of $[\log \alpha_{1\cdot2} - \log (p_1^\circ/p_2^\circ)]$ against x_1 should yield a smooth curve, and this property can provide a rough check on the accuracy of the data when cast into a form suitable for the above test.

For the systems marked * in the eighth column of Table III the data are probably inconsistent, *i.e.*, for these systems $(D–J) > 10$.

TABLE III

Analysis of Data from Perry's Handbook [25]

Components		Pressure, mm Hg	θ	T_i	Σ	D	J
Acetic acid	Benzene	760	38·0	353·2	0·1998	57·0	16·1 *
Acetic acid	Water	760	18·1	373·2	0·0528	100·0	7·3 *
Acetone	Ethanol	760	21·9	329·7	0·1420	40·2	10·0 *
Acetone	Methanol	760	8·2	329·7	0·1269	12·2	3·7
Benzene	Ethanol	750	11·1	341·0	0·3287	3·2	4·8
n-Butanol	Water	760	19·3	365·9	0·4316	2·0	7·9
i-Butanol	Water	760	18·8	362·4	0·4092	0·3	7·8
Carbon tetrachloride	Benzene	760	3·4	349·9	0·0195	10·8	1·5
Carbon tetrachloride	Ethyl acetate	685	2·4	344·8	0·0773	21·3	1·0 *
Carbon tetrachloride	Toluene	762	33·1	350·9	0·0542	100·0	13·3 *
Carbon disulphide	Carbon tetrachloride	760	30·2	319·7	0·0551	38·9	14·3 *
Carbon disulphide	Acetone	760	17·4	312·3	0·3079	5·2	8·4
Chloroform	Acetone	760	7·9	329·7	0·1449	11·8	3·6
Chloroform	Benzene	760	18·8	334·5	0·0850	22·8	8·4 *
Ethanol	Water	760	21·8	351·4	0·2544	4·8	9·3
Ethyl acetate	Ethanol	760	6·6	345·0	0·1718	2·7	2·9
Ethylene glycol	Water	228	94·2	342·5	0·0727	71·2	41·2 *
Furfural	Water	760	63·9	371·1	0·4670	26·5	25·7
Methanol	Water	760	35·3	337·9	0·1494	1·9	15·7
Nitrogen	Oxygen	760	13·1	77·5	0·0406	22·7	25·4
Nitrogen	Oxygen	3800	15·0	94·2	0·0655	100·0	23·8 *
*iso*Propyl ether	*iso*Propanol	760	16·3	339·4	0·2597	13·3	7·2
*iso*Propanol	Water	760	19·5	353·7	0·4026	12·6	8·3
n-Propanol	Water	760	12·2	361·0	0·3386	11·1	5·1

ACTUAL EXAMPLES OF THE EFFECT OF NON-IDEALITY OF VAPOURS ON THE TEST

The derivation of the second virial coefficient from experimental measurements is difficult (see, for example, Hirschfelder, McClure, and Weeks [30] for a discussion), and consequently this quantity has frequently been calculated from values of the second virial coefficient of water using the principle of corresponding states (*e.g.*, see Scatchard and Raymond,[9] Gordon and Hornibrook [31]). This procedure, however, often yields calculated values differing from the observed by a factor of as much as two or more; so that this method will not be adopted here, and only experimentally observed values of the virial coefficients will be used.

The results of the computation of the effect of deviations from the gas laws are summarized in Table IV for those systems given in Table III for which B_{11} and B_{22} values are available.

The values of Z in Table IV were obtained by means of the approximate relationship (25) and D (corrected) calculated by equation (24). Comparison of columns 10 and 11, Table IV, shows that Z is not numerically large, and only in the case of the system carbon tetrachloride–benzene, where Σ is very small, does the correction arising from Z modify the conclusion concerning the consistency of the data. It may be noted that the value of

TABLE IV

The Effect of Deviations from the Gas Laws on the Test

Component 1	Component 2	$-B_{11}$	Ref B_{11}	$-B_{22}$	Ref B_{22}	Σ	I	Z	D	D corrected	J
Acetone	Methanol	1360	32	1030	33	0·1269	−0·0155	+0·0052	12·2	8·3	3·7
Carbon tetrachloride	Benzene	1050	32	1160	33	0·0195	+0·0021	+0·0016	10·8	19·0 *	1·5
Chloroform	Acetone	840	32	1360	32	0·1449	−0·0171	−0·0032	11·8	14·0	3·6
Chloroform	Benzene	840	32	1160	33	0·0850	+0·0194	+0·0100	22·8 *	34·6 *	8·4
Methanol	Water	1030	33	452	33	0·1494	−0·0029	+0·0122	1·9	6·2	15·7

Z for the system methanol–water is only 0·012, although both the virial coefficients and the vapour pressures of the components differ considerably.

Unfortunately experimental values of the second virial coefficient for many of the materials listed in Table III are not available, but in view of the Z values given in Table IV and of the Σ values in Table III it is improbable that the application of the corrections for deviation from the gas laws to the data in Table III would seriously modify the conclusions reached on the consistency of the data on the majority of these systems. Possible exceptions, however, are the systems carbon tetrachloride–ethyl acetate and carbon disulphide–carbon tetrachloride, where the effect of deviations from the gas laws might be just sufficient to reverse the decision on the inconsistency of the data.

ACKNOWLEDGMENTS

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List of Symbols

x_1 and x_2	mol fractions of components 1 and 2 in the liquid respectively.
y_1 and y_2	mol fractions of components 1 and 2 in vapour respectively.
$\alpha_{1\cdot2}$	relative volatility of component 1 with respect to component 2; $(y_1/x_1)(x_2/y_2)$.
γ_1 and γ_2	activity coefficients of component 1 and 2 with respect to the pure liquids as standard states and with compositions expressed as mol fractions.
p_1° and p_2°	vapour pressures of pure components 1 and 2.
ln	logarithm to base e.
log	logarithm to base 10.
I	$\int_0^1 [\log \alpha_{1\cdot2} - \log (p_1^\circ/p_2^\circ)]dx_1$.
$\lvert I \rvert$	value of I taken with a positive sign.
Σ	total area on the plot of $[\log \alpha_{1\cdot2} - \log (p_1^\circ/p_2^\circ)]$ against x_1 taken with a positive sign where no cognizance is taken of the sign of the individual areas (*i.e.*, area A plus area B, Fig. 1).
D	percentage deviation; $(100\lvert I \rvert)/\Sigma$.
θ	boiling range of isobaric data.
$\lvert\theta\rvert$	value of θ taken with a positive sign.
T_i	lowest boiling point (° K) in isobaric data.
t_i	lowest boiling point (° C) in isobaric data.
J	$[150\lvert\theta\rvert/T_i]$.
ΔG^E	excess free energy of mixing.
ΔH	heat of mixing.
ΔS^E	excess entropy of mixing.
T_1° and T_2°	boiling points (° K) of pure components 1 and 2 under the isobaric pressure.
R	gas constant; 82·06 cm³ atm/deg. mol.
$\lvert \Delta H_m \rvert$	numerically largest value of ΔH taken with positive sign which occurs in concentration range $x_1 = 0$ to 1.

$\lvert\Delta G_m{}^E\rvert$	numerically largest value of ΔG^E taken with positive sign which occurs in concentration range $x_1 = 0$ to 1.
V_1 and V_2	molar volumes of vapours of pure components 1 and 2.
$\bar{V}_1$ and $\bar{V}_2$	partial molar volumes of the vapours of components 1 and 2 in the vapour mixture.
P	total pressure of vapour.
α_1	$(RT/P) - V_1$.
α_2	$(RT/P) - V_2$.
$\bar{\alpha}_1$	$(RT/P) - \bar{V}_1$.
$\bar{\alpha}_2$	$(RT/P) - \bar{V}_2$.
B_{11} and B_{22}	second virial coefficients of pure components 1 and 2.
B_{12}	coefficient in equation (18).
Z	$(1/2{\cdot}303)\int_0^1 (1/RT)\,[B_{11}(P - p_1^\circ) - B_{22}(P - p_2^\circ)]\,dx_1$.
$\lvert I + Z\rvert$	Algebraic sum of I and Z taken with positive sign.
D (corrected)	$(100\lvert I + Z\rvert)/\Sigma$.
$T_{\text{azeo. (min)}}$	boiling point (° K) of minimum-boiling azeotrope.
$T_{\text{azeo. (max)}}$	boiling point (° K) of maximum-boiling azeotrope.

Note the difference between $\alpha_{1\cdot2}$, which is a volatility ratio, and α_1, which is the volume residual quantity defined above.

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HYDRODYNAMIC LUBRICATION OF ROTATING DISKS IN PURE SLIDING. A NEW TYPE OF OIL FILM FORMATION

By A. CAMERON *

SUMMARY

Reynolds' classical hydrodynamic lubrication theory leads to equations which show that the formation of an oil film between disks is dependent on the rolling velocity only.

Experiments have been carried out by the author which show that it is possible to get an oil film under conditions of pure sliding with no rolling at all. Classical lubrication theory will not explain this new effect.

The explanation advanced for it is that the variation of viscosity across the thickness of the film introduces an extra term into the standard equation of film lubrication. Making two simplifying assumptions for the temperature of the surfaces in contact and the temperature distribution in the oil film, the theory is tested numerically.

NOTATION

a = constant in the oil viscosity–temperature equation.
C = compressibility of oil.
E = Young's Modulus.
F = shear force.
h = film thickness at any point.
$\bar{h}$ = film thickness when $dp/dx = 0$.
h_0 = minimum film thickness.
$h_1 h_2$ = film thickness on either side of temperature peak.
k = constant in the oil viscosity–temperature equation.
K = thermal conductivity.
m = oil viscosity–temperature coefficient.
M = constant in variable viscosity equation.
p = pressure at any point.
q = heat flux at the surface.
Q = rate of oil flow.
R = reduced radius $R = \dfrac{R_1 \cdot R_2}{R_1 + R_2}$.
T = " viscosity " temperature at any point.
T_1 = " viscosity " temperature of cold disk.
t_1 = actual temperature rise (maximum).
t = actual temperature.
v = velocity of a particle of lubricant in the x-direction of motion.
$V_{1\text{-}2}$ = surface velocity of disks 1, 2.
V_R = rolling velocity $V_R = V_1 + V_2$.
V_S = sliding velocity $V_S = V_1 - V_2$.
V = volume.
w = elastic deflexion of surface.
W = load per unit length.
x = co-ordinate in the direction of motion.
y = co-ordinate through the film.
z = co-ordinate along the axis.
α = h_1/h_2, where $0 \leq \alpha \leq 1$.
η = viscosity.
θ = relative temperature rise t_1/T_1.
Θ = constant in variable viscosity equation.
κ = thermal diffusivity.
ρ = density of oil.
ξ = pressure–viscosity coefficient.

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Introduction

While there is a reasonably large amount of agreement between theory and practice in the lubrication of bearings, gearing results have so far defied theoretical attack. The work described here is part of a programme designed to apply theory to practical results, and at the outset of this programme of work the phenomenon described here was observed. Briefly, the lubrication of gears—or disks to which they are kinematically similar—should depend only on their rolling velocity, and not at all on their sliding velocities (these terms are defined later). Experiment showed that the sliding velocity of disks plays a very important part in their lubrication, in fact, disks could carry a sizable load on an oil film produced by pure sliding, with no rolling at all.

Experimental Method

The machine used for these tests is in essence the same as the one described by Merritt.[8]

It consists of two disks, 4 inches and 8 inches in diameter, making contact along a line $\frac{3}{4}$ inch long and loaded by carrying one disk in a swinging frame and pressing it into contact with the other by a lever and jockey weight. The 8-inch fixed-axis disk is driven through suitable gearing by a variable-speed motor mounted in a swinging cradle, constrained by a spring balance. The torque on the motor can be determined from the pull on the spring balance, which is calibrated in pound-inches. The 4-inch swinging-axis disk is driven from the 8-inch disk.

In these experiments pure rolling and pure sliding were required. For pure rolling the 4-inch disk was allowed to run free, being driven by friction from the 8-inch disk. No measurable slip was found. The pure sliding condition was achieved by fixing sprockets on the 8- and 4-inch disk shafts of 48 and 24 teeth respectively, and connecting them by chain. This gave the 4-inch disk double the *angular* velocity of the 8-inch disk. The peripheries of the two disks had surface speeds of the same magnitude but of opposite sign, *i.e.*, at the point of contact there was pure sliding.

There was an electric heater in the oil-bath to bring the oil to the operating temperature, which was normally 160° F. A small jack was fitted which, when screwed down, lifted the swinging frame so that the two disks were out of contact. When the disks were out of contact the fractional losses of the bearings and chains and the churning losses of the oil could be determined over the desired range of speeds. These were subtracted from the torque found when the disks were loaded together. The no-load losses served as a useful check to see that the machine was running smoothly. If the readings were abnormally high it usually indicated incorrect assembly of the disks, chain, or gearing from the motor to the machine, thus preventing a whole run being spoilt.

The 8-inch disk normally consisted of a centrifugally cast bronze rim shrunk and pinned on to a 5-inch cast-iron blank. This itself was bored and keyed on to the shaft.

In some of the experiments described below the electrical conductivity of the oil film was measured. To enable this to be done the bronze disk was mounted on a 3-inch fabric plastic (Tufnol) boss. An insulated wire

was led from the bronze rim, through a hole bored axially in the shaft to an insulated brass thimble mounted on the stub shaft end. A spring-loaded carbon brush was used as collector. A ball bearing was tried initially, but was found to have too high a resistance when rotating. The return circuit was through the metal casing of the machine. The resistance between the casing and the 4-inch disk was found to be negligible.

The electrical resistance was measured by a standard Avometer which contains a 4½-volt battery in series with a milliameter. A high voltage was used to ensure that the oil film would have to be properly formed before it could withstand the applied voltage without electrical breakdown (see also Cameron [5]).

The viscosity of the oil used throughout these experiments is shown in Table I.

TABLE I

Temperature, ° F	Viscosity, cs
70	1000
100	330
140	100
200	28
Density at 60° F = 0·895	

THEORY

(a) *Constant Viscosity*

The load-carrying capacity of disks using the assumption of constant viscosity has been obtained a number of times. [1, 6, 7, 9, 12] The derivation briefly is as follows :—

The basic equation of all hydrodynamic lubrication with constant or variable viscosity and neglecting side leakage is :—

$$\frac{\partial F}{\partial y} = \frac{dp}{dx} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad \text{(i)}$$

where F = shear force, p = pressure, and x and y co-ordinates are respectively parallel and perpendicular to the direction of motion.

Newton's law of viscous drag states that

$$F = \eta \frac{\partial v}{\partial y} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad \text{(ii)}$$

where η = viscosity and v = velocity in the x-direction of any particle of lubricant

thence

$$\frac{dp}{dx} = \frac{\partial}{\partial y}\left(\eta \frac{\partial v}{\partial y}\right) = \eta \frac{\partial^2 v}{\partial y^2} + \frac{\partial \eta}{\partial y} \cdot \frac{\partial v}{\partial y} \quad . \quad . \quad . \quad . \quad . \quad . \quad \text{(iii)}$$

It is usual to neglect the variation of viscosity across the film, and hence the standard equation becomes, as in Reynolds' original theory :—

$$\frac{dp}{dx} = \eta \frac{\partial^2 v}{\partial y^2} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad \text{(iv)}$$

If V_1 and V_2 are the velocities of the surfaces (positive in x-direction) and neglecting side leakage, which in this case is justifiable, then :—

$$\frac{dp}{dx} = 6(V_1 + V_2)\eta\left(\frac{h - \bar{h}}{h^3}\right) \quad . \quad . \quad . \quad . \quad . \quad . \quad \text{(v)}$$

where h is the film thickness at any point x and $\bar{h}$ is the film thickness when $dp/dx = 0$. It is seen that if $V_1 = -V_2$, then dp/dx is always zero, and hence p is uniformly nought. This conclusion is the same if the lubricant is considered compressible or if it expands with any increases in temperature that may occur along the x axis (thermal wedge effect).

Equation (v) is integrated to give the pressure curve for a pair of disks, and this curve again can be integrated to give the total load carried by the oil film (see authors cited). This is :—

$$W = 2{\cdot}45\ (V_1 + V_2)\eta \cdot \frac{R}{h_0} \quad . \quad . \quad . \quad . \quad . \quad . \quad \text{(vi)}$$

where h_0 is the minimum film thickness.

We will now consider the variable viscosity condition.

(b) *Variable Viscosity*

(i) *General Theory.*—The fact that the viscosity is not constant across the oil film may allow a sliding-velocity term to appear in the final load equations (a suggestion made to the author by Prof. C. Weber).

The starting equation is equation (iii)

$$\frac{dp}{dx} = \eta\frac{\partial^2 v}{\partial y^2} + \frac{\partial \eta}{\partial y} \cdot \frac{\partial v}{\partial y}.$$

The solution of this differential equation involves the solution of a number of unknown problems.

The following variables should be taken into account :—

(1) Variation of viscosity of the oil film with temperature and pressure, in both x and y directions.

(2) Surface temperature of the teeth due to the frictional heat being conducted away through them. This will give the boundary temperatures for (1).

(3) Elastic deformation of the surfaces by the pressure.

(4) Adiabatic heating of the oil. This is about 1° C per 100 atmospheres.[2b] Pressures of the order of 2000–4000 atmospheres are reached in an oil film between gear-teeth.

In the Appendix the differential equations for these varying effects are set up. Work is being carried out on their solution elsewhere, and will be published in time.

(ii) *Simplified Theory.*—These differential equations, taking into account the four variables listed above, are so complex that no immediate solution of them can be expected. As an analysis of the experimental results presented here, two large simplifications have been made to enable preliminary calculations to be made.

It is assumed :—

(i) that one disk is uniformly hotter than the other, and

(ii) that the temperature in the oil film follows a known law.

and that there are two types of temperature distribution :—

(1) linear temperature drop from the hot to the cold disk, Fig. *a*;

(2) parabolic distribution, allowing the maximum to occur anywhere in the oil film, not necessarily at the surface, Fig. *b*.

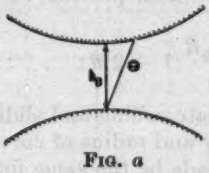

FIG. *a*

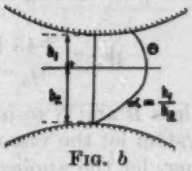

FIG. *b*

In the latter parabolic distribution are defined $\alpha = h_1/h_2$, where h_1 and h_2 are the film thicknesses on either side of the temperature maximum; h_1 and h_2 make up the total film thickness at any point. It is seen that if $h_1 = h_2$, the temperature distribution is symmetrical and $\alpha = 1$, if $h_1 = 0$, $\alpha = 0$, the maximum is on the top surface.

To calculate the variation of viscosity with temperature we will use a formula due to Slotte [11] in place of the usual logarithmic temperature viscosity relation of the Walther or Vogel type.[2a]

Slotte's formula can be written :—

$$\eta = \frac{k}{(t+a)^m} = \frac{k}{T^m}$$

where η = viscosity at any temperature, m, k, and a = constants for a given oil, and T is defined as $(t + a)$.

If t is in ° Centigrade, then $a \simeq 40$–50, hence we can define a Centigrade " viscosity–temperature " scale starting at $-a$° Centigrade, which gives a simple reciprocal power–temperature law.

The oil used in these experiments had a value of $a = 46$ and $m = 5$.

The equations can now be integrated and their form seen.

This is done in Appendix II, using the assumptions given above, and the load carried is :—

$$W = \frac{4 \cdot 9}{\Theta}\{(V_1 - V_2)\,M + V_2\}\frac{\eta_0 R}{h_0} \quad . \quad . \quad . \quad \text{(vii)}$$

where Θ and M are constants; η_0 = viscosity at the cold disk; other notation as before.

Now this equation is seen to differ from the previous constant-viscosity equation (vi) by the addition of the two new constants Θ and M. A study of their derivation shows that Θ is a factor which, when divided into the " base " or cold disk viscosity, gives the mean effective viscosity. M, on the other hand, allows for the distortion of the stream lines.

It can be seen from the equations that for constant viscosity, ($m = 0$), or zero temperature rise ($\theta = 0$), M becomes equal to $\frac{1}{2}$ and Θ becomes 1. Thus equation (vii) goes over into the constant–viscosity equation (vi), the expression :—

$$\{(V_1 - V_2)\,M + V_2\} \text{ becoming } \frac{V_1 + V_2}{2}.$$

If there is pure rolling, *i.e.*, $V_1 = V_2$, then equation (vii) becomes, putting $V_R = V_1 + V_2$,

$$W_R = \frac{2{\cdot}45}{\Theta_R}\frac{V_R\eta_0 R}{h_0} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad \text{(viii)}$$

and where there is pure sliding it becomes, putting $V_S = (V_1 - V_2)$ and $V_1 = -V_2$,

$$W_S = \frac{2{\cdot}45}{\Theta_S}\frac{V_S\eta_0 R}{h_0}(1 - 2M) \quad . \quad . \quad . \quad . \quad . \quad . \quad \text{(ix)}$$

using the subscripts R and S to indicate rolling and sliding respectively.

For a given system let the viscosity and radius of curvature of the disks be fixed. Further, let the applied loads be the same for both rolling and sliding, *i.e.*, let $W_R = W_S$. If some method can be used such as electrical

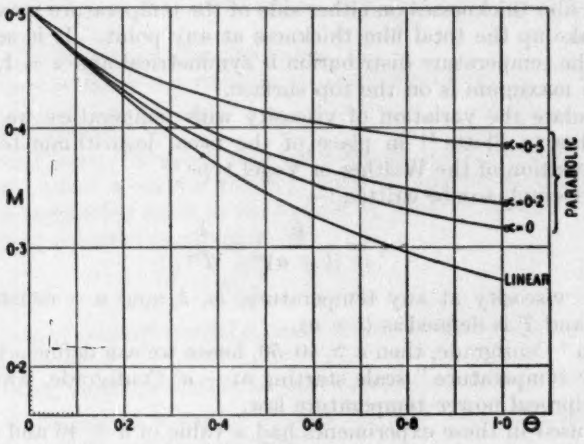

FIG. 1

SLIDING DISKS. VARIATION OF M WITH θ

resistance, to gauge when the thickness of the oil film is the same in both cases, then the ratio of the rolling to sliding velocities required to produce equal oil-film thickness is given from equations (viii) and (ix) :—

$$\frac{V_R}{V_S} = (1 - 2M)\frac{\Theta_R}{\Theta_S} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad \text{(x)}$$

Computed Results

Now the sizes of M and Θ are dependent on the temperature–viscosity characteristics of the oil, the relative temperature rise θ, and in the case of the parabolic temperature distribution, α (α is defined from Fig. *b supra*).

The quantities M and Θ have been computed, and are displayed in Figs. 1 and 2 for values of θ between 0 and 1 with $m = 5$ for both the linear temperature distribution and the parabolic distribution with $\alpha = 0, 0{\cdot}2$, and $0{\cdot}5$.

A reasonable approximation to Θ, in the case of $m = 5$ and the parabolic distribution, is found to be given by $\Theta = (1 + \frac{2}{3}\theta)^5$. The same approximate

relation was given by Cameron and Wood [3] for the variation of viscosity in the direction of motion for the thermal wedge bearing. For values of m other than 5, this same approximation, $\Theta = (1 + \frac{3}{5}\theta)^m$, is used to determine Θ. Values of M are computed from the full equation, as it is quite short.

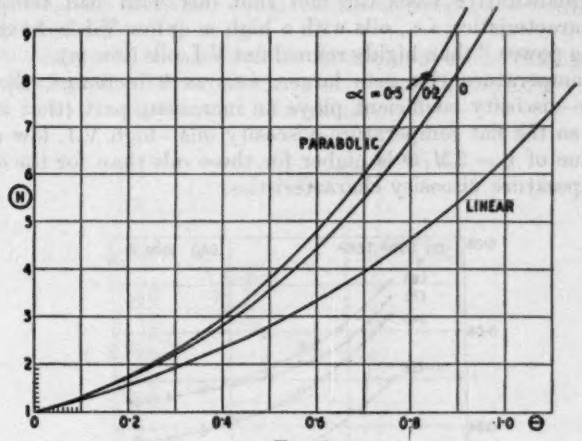

FIG. 2

SLIDING DISKS. VARIATION OF Θ WITH θ

In the case of the linear temperature distribution, a very good approximation of $m = 5$ is given by $\Theta = (1 + 0{\cdot}450)^5$. A series of trial calculations at $\theta = 1$ shows that this holds reasonably well for all values of m between 1 and 15.

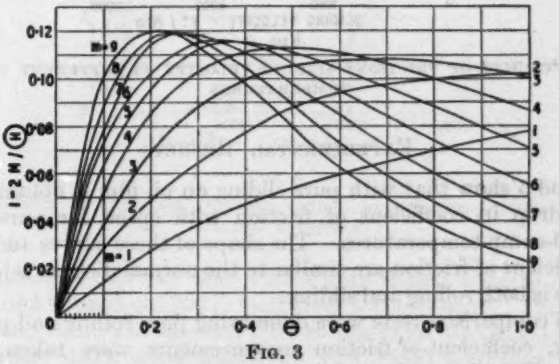

FIG. 3

SLIDING DISKS. VALUE OF $1-2M/\Theta \sim \theta$ FOR DIFFERENT m.
$\Theta = (1 + 0{\cdot}45\,\theta)m$

The load-carrying capacity under pure sliding, from equation (ix), is proportional to $\left\{\frac{1-2M}{\Theta}\right\}_g$ for any given system. This function is plotted in Fig. 3 for a range of values of θ, the non-dimensional temperature rise, and of m the temperature–viscosity coefficient of the oil.

As a slight digression it may be noted that Fig. 3 gives an explanation of an effect that has been often noticed but not previously explained. Looking at the figure it is seen that at low values of θ (small temperature rise) the values of $(1 - 2M)/\Theta$ is higher the larger the value of m. This explains on a semi-quantitative basis the fact that oils with bad temperature–viscosity characteristics, *i.e.*, oils with a high m or low V.I.'s, have a better " lubricating power " than highly refined flat V.I. oils (low m).

As the temperature rise gets larger, *i.e.*, as θ increases, the normal temperature–viscosity coefficient plays an increasing part (that is, Θ gets larger) and so the flat temperature–viscosity oils—high V.I. low m—score and the value of $1 - 2M/\Theta$ is higher for these oils than for the ones with steeper temperature viscosity characteristics.

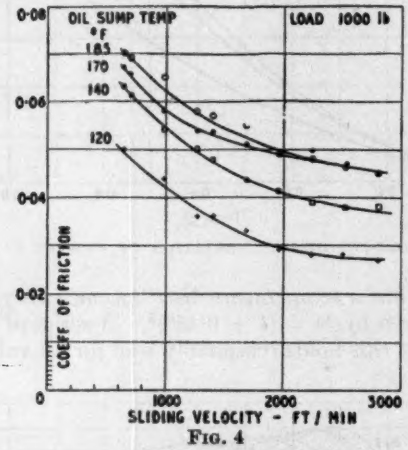

FIG. 4

COEFFICIENT OF FRICTION—SLIDING VELOCITY AT DIFFERENT SUMP TEMPERATURES

EXPERIMENTAL RESULTS

Figs. 4 and 5 show that with pure sliding an oil film is obtained. Fig. 4 gives the drop in coefficient of friction with speed for pure sliding at different oil-sump temperatures. The shape of these curves and the values of the coefficient of friction are similar to the normal disk tests (cf. Merritt), where there is both rolling and sliding.

In Fig. 5 comparison tests were done using pure rolling and pure sliding. For sliding, coefficient-of-friction measurements were taken. The low value of 0·015 which was reached shows that there must have been full fluid lubrication. The electrical resistance was measured in both cases.

If Fig. 5 is studied it will be seen that the two electrical-resistance curves are of the same shape. In fact, for any given resistance, the ratio between the sliding and rolling speeds is practically constant. This would seem to show that the oil film in each case must *act* in the same way (though it is not formed in the same manner) on the disk surfaces, and the process of the change from boundary to full fluid lubrication is the same.

The same comparison experiments were carried out at an oil-sump temperature of 140° F, and ratio V_R/V_S was almost constant at 0·75.

The electrical resistance of the oil film was used as a means of gauging the thickness of the film, it being assumed that for any given load, oil viscosity, surface finish, and material, equal electrical resistances corresponded to equal film thicknesses.

This would seem to be a reasonable assumption, as the rolling and sliding speeds for any given oil-film resistance have a sensibly constant ratio, one to the other, over the entire range of resistance.

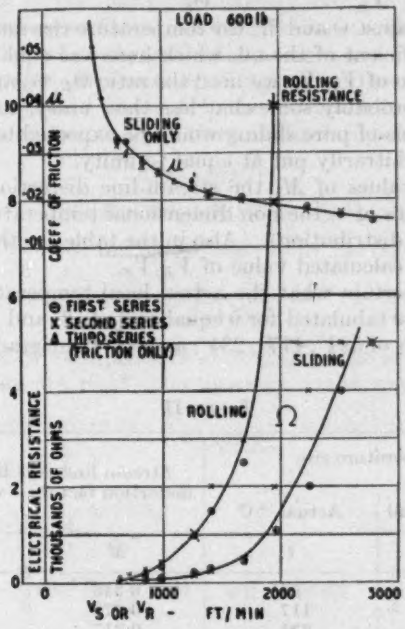

FIG. 5

COEFFICIENT OF FRICTION AND ELECTRICAL RESISTANCE—SLIDING AND ROLLING VELOCITY

It was noticed in this test that with a pure sliding velocity of 420 feet/minute, the disks seized at 1200 lb load (over a $\frac{3}{4}$-inch face). Previous work had shown that if, instead of using pure sliding there was equal sliding and rolling (*i.e.*, the disks being joined by 1 : 1 chain sprockets giving the *same angular* velocity to each), then the seizure load would have been some 4000 lb. Pure sliding, therefore, does not form such a good film as mixed sliding and rolling. Running a pair of case-hardened steel disks against each other produced almost immediate scuffing. This is not surprising, as the boundary lubrication—or mixed friction-running properties of steel on steel—are much worse than those of steel–bronze. Hence any momentary touching of the surfaces under these extreme conditions of sliding would cause the surfaces to tear and scuff.

A further point is that with the steel–steel combination the thermal properties of the two surfaces are identical, hence the thermal asymmetry necessary to form the oil film will not be produced so readily. The exact influence of the different thermal properties of the surfaces should appear from the full theoretical analysis.

Comparison of Theory and Experiments

In the theoretical section we have obtained a formula for the ratio V_R/V_S, equation (x), this is $\frac{V_R}{V_S} = (1 - 2M)\frac{\Theta_R}{\Theta_S}$. Values of M and Θ are plotted in Figs. 1 and 2, against Θ and M, the temperature rise and M, the temperature–viscosity coefficient of the oil, which here was equal to 5.

To obtain a value of V_R/V_S we need the ratio Θ_R/Θ_S and the value of M. Now Θ_R/Θ_S is probably somewhat less than unity, as the temperature rise under conditions of pure sliding would be expected to be larger than in rolling. This is arbitrarily put at equal to unity.

Table II gives values of M, the stream-line distortion factor obtained from different values of θ, the non-dimensional temperature rise (using the linear temperature distribution). Also in the table are the actual temperature rise t and the calculated value of V_R/V_S.

It is not at all certain what the actual local temperatures of disks are. Therefore values are tabulated for θ equal to $\frac{1}{2}$, 1, 2, and ∞, corresponding to temperature rises of $58\frac{1}{2}°$, 117°, 234°, and $\infty°$ Centigrade.

Table II

Temperature rise		Stream line distortion factor	Rolling/sliding velocity ratio
Non-dimensional	Actual, ° C		
θ	t	M	V_R/V_S
$\frac{1}{2}$	$58\frac{1}{2}$	0·346	0·308
1	117	0·272	0·456
2	234	0·217	0·566
∞	∞	$\frac{1}{m+2} = 0{\cdot}143$	0·714

The observed value of V_R/V_S is 0·7 to 0·75. The calculated figures are 40 to 80 per cent of the true values. Considering the very large assumptions made, the agreement is as good as can be expected. It shows that the mechanism put forward to explain the new phenomenon gives results which harmonize tolerably well with experiments.

Conclusions

In conclusion, it is seen that this simple theory explains semi-quantitatively the results observed with sliding disks. The fact that the agreement is not exact is only to be expected from the assumptions made that one disk is uniformly hotter than the other, and that there is a regular temperature

distribution in the oil film. Work on the accurate solution is now being undertaken, and it is hoped that the quantitative figures will be in much greater harmony with experiment. This work will also take into account the effect of pressure on viscosity and of deformation of the flanks, factors which are important in gear lubrication.

Future work on lubrication generally will have to consider the variation of viscosity across the film, which up to now has, with no detriment, been ignored.

Acknowledgments

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Appendix I

Full Hydrodynamic Equation

The oil pressure between the flanks is given by the equation :—

$$\frac{dp}{dx} = \frac{\partial}{\partial y}\left\{\eta \frac{\partial y}{\partial y}\right\}.$$

The viscosity varies with pressure according to the law $\eta = \eta_0 \exp(\xi p)$. Assuming the flanks are rigid—the viscosity varies only with pressure—we get, using the substitution $\tan\psi = \dfrac{x}{\sqrt{2Rh_0}}$ (see authors cited) :—

where $\bar{\psi}$ corresponds to $\bar{h}$

$$\frac{1 - e^{-\xi p}}{\xi} = 3(V_1 + V_2)\,\eta_0 \frac{\sqrt{2Rh_0}}{h_0^2} \left[\left(1 - \frac{3}{4}\sec^2\bar{\psi}\right)\left(\frac{\pi}{2} + \psi + \sin\psi\cos\psi\right) - \frac{1}{2}\sec^2\bar{\psi}\sin\psi\cos^3\psi\right]$$

and

$$1 - e^{-\xi p_{max}} = 0{\cdot}76\,\eta_0 \xi (V_1 + V_2)\sqrt{\frac{2R}{h_0^3}}$$

we see that $p_{max} \longrightarrow \infty$ when the R.H.S. $\longrightarrow 1$.

The load carried is $\int_a^b p\,dx$, where a and b are the limits of the pressure curve. These are given from the Reynolds boundary conditions (Cameron and Wood [4]).

It can be shown that when the R.H.S. $= 1$ this integral is still finite. If there is now an increase of speed the R.H.S. cannot be greater than 1, so the film thickness h_0 must increase (assuming ξ and η_0 are constant). Thus the disks become oil-borne whatever the load. A preliminary study of this subject is in the press (Cameron [5]).

The total heat production is made up of two terms, the normal viscous drag and the adiabatic heating. This heat is removed by convection—that is the heating up of the oil—and conduction through the oil film.

It can be assumed that conduction lengthwise along the oil film (in the x direction) is small.

The equations are as follows :—

$$C = \text{compressibility}$$

$$= \frac{1}{V}\frac{dV}{dp}, \text{ where } V = \text{volume} = \frac{1}{\rho}$$

$$= \frac{\rho}{dp/dx}\frac{d}{dx}\left\{\frac{1}{\rho}\right\}$$

where ρ = density; p = pressure; v = velocity of oil particle.

Then the heat produced by compression

$$= \iiint_V p\frac{\partial v}{\partial x}\,dx\,dy\,dz$$

$$= \iiint_V p\rho\frac{\partial}{\partial x}\left\{\frac{1}{\rho}\right\}dx\,dy\,dz \text{ (see Ramsey, "Hydrodynamics," p. 376)}$$

$$= C\iiint_V pv\frac{dp}{dx}\,.\,dx\,dy\,dz$$

where η = viscosity; c = specific heat of oil; K = thermal conductivity; t = temperature; f = heat flux.

Heat produced by viscosity losses

$$= \eta\iiint_V \left\{\frac{\partial v}{\partial y}\right\}^2 dx\,dy\,dz.$$

Heat gained by convection

$$= \iiint_V \left\{-c\rho v\frac{\partial t}{\partial y}\right\}dx\,dy\,dz.$$

Heat gained by conduction

$$= \iiint_V \frac{\partial}{\partial y}\{f_y\}dx\,dy\,dz$$

$$= \iiint_V K_{\text{oil}}\frac{\partial^2 t}{\partial y^2}\,dx\,dy\,dz, \text{ since } f_y = K_{\text{oil}}\frac{\partial t}{\partial y}.$$

Hence equating heat gained and lost, and since the integration is true for any volume in the region

$$\eta\left\{\frac{\partial v}{\partial y}\right\}^2 + Cpv\frac{dp}{dx} + c\rho v\frac{\partial t}{\partial x} - K_{\text{oil}}\frac{\partial^2 t}{\partial y^2} = 0.$$

The flow of heat through the metal surface gives the value of the boundary temperature of the disks.

The temperature rise t at a point x due to a flow of heat q, where $q = (f_y)\,y = h$ over an infinitesimal distance dx_1 at x_1 is given by the relation

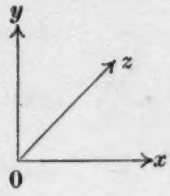
$$t_1 = \frac{1}{\pi K_{\text{metal}}}\int_a^b qe^{V(x-x_1)/2\kappa}K_0\{V(x-x_1)/2\kappa\}dx_1$$

where K_{metal} = conductivity of the metal surface; V = surface velocity (not volume); κ = diffusivity of metal; a and b = extent of the pressure curve or oil film; $K_0(x)$ = modified Bessel function of the second kind of order zero. Cf. H. S. Carslaw and J. C. Jaeger, "Conduction of Heat in Solids" (Eqn. 4, p. 107), Oxford, 1947.

The value of q is obtained from the heat generated in the oil film.

The Distortion of the Disk Surfaces

The deflexion w of a surface (x, o, z) due to a pressure p acting over a small area $(dx_1\, dz_1)$ at the point (x_1, o, z_1) is



$$w = -\frac{1-\nu^2}{\pi E}\phi$$

where ν = Poisson's ratio; E = Young's modulus. Cf. A. E. H. Love, "The Mathematical Theory of Elasticity," Art. 138. 4th Edn. Cambridge, 1927.

$$\phi = \iint \frac{p}{r} dz_1\, dx_1$$

$$r = \sqrt{(x-x_1)^2 + y^2 + (z-z_1)^2}.$$

We are interested in the strip from l to $-l$ in the z direction and from a to b (the extent of the pressure curve), in the x direction, and we will let $l \longrightarrow \infty$. Thus w is independent of z, and we can therefore consider the deflexion at the origin and put $z = 0$.

Let $$e^2 = (x - x_1)^2 + y^2$$

then $$\phi = \int_a^b \int_{-l}^{l} \frac{p dz_1 dx_1}{\sqrt{e^2 + {z_1}^2}}$$

$$\int_{-l}^{l} \frac{p dz_1}{\sqrt{e^2 + {z_1}^2}} = 2p \log\left\{\frac{l + \sqrt{e^2 + l^2}}{e}\right\}$$

now $e^2/l^2 < < 1$

$$\text{then} \int_{-l}^{l} \frac{p dz_1}{\sqrt{e^2 + {z_1}^2}} = 2p \log \frac{2l}{e} = 2p \log 2l - 2p \log e$$

$$\text{so as } l \longrightarrow \infty, \quad = -p \log l^2 + \text{infinite constant.}$$

This infinite constant may be ignored, as we are dealing with changes in w. On the surface $y = 0$ the total distortion

$$w = w_1 + w_2 = -\left[\frac{1-{\nu_1}^2}{\pi E_1}\right]\phi_1 - \left[\frac{1-{\nu_2}^2}{\pi E_2}\right]\phi_2$$

if both flanks are of the same material

$$w = -\frac{2(1-\nu^2)}{\pi E}\int_a^b p \log (x_1 - x)^2\, dx_1$$

where a and b denote the extent of the pressure curve.

This can also be obtained in non-dimensional form if p is put equal to p^1E and all lengths are referred to h_0, the minimum film thickness,

then $$\frac{w}{h_0} = w^1 = -\frac{2(1-\nu^2)}{\pi}\int_{a^1}^{b^1} p^1 \log (x_1{}^1 - x^1)^2\, dx_1{}^1$$

Primes show non-dimensional quantities.

Appendix II

(a) *Linear Temperature Distribution*

Temperature T at y is $T = T_1 + \frac{y}{h}t_1$

$$\eta = \frac{k}{T^m}$$

$$= \frac{k}{T_1{}^m\left(1 + \frac{yt_1}{hT_1}\right)^m}$$

$$= \frac{\eta_0}{\left(1 + \frac{y}{h}\theta\right)^m}$$

where T_1 = temperature of cold disk; t_1 = temperature rise (maximum); h = film thickness; η = viscosity; k and m constants for the oil; T = " viscosity " temperature (see text) = (° C+ a constant, about 40); η_0 = viscosity of oil at T_1; θ = relative temperature rise.

$$\frac{dp}{dx} = \frac{\partial}{\partial y}\left(\eta\frac{\partial u}{\partial y}\right) \text{ from equation (iii) in text.}$$

$$\therefore \quad u = \frac{s^{m+2}h^2}{(m+2)\theta^2\eta_0}\frac{dp}{dx} + C_1\frac{s^{m+1}}{m+1} + C_2$$

where $$s = \left(1 + \theta\frac{y}{h}\right)$$

C_1 and C_2 integration constants.

The oil flow $$Q = \int_0^h (u\,dy).$$

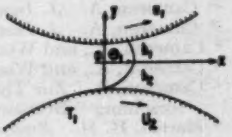
Hence

$$Q = \frac{h^3}{\eta_0}\frac{dp}{dx}\left\{\left\{\frac{\theta^2(m+2)^2(1+\theta)^{m+1} - \{(1+\theta)^{m+2} - 1\}^2}{(m+2)^2(m+3)\theta^3(1+\theta)^{m+1} - 1}\right\}\right\}$$
$$+ h\left[(U_1 - U_2)\left(\frac{(1+\theta)^{m+2} - 1}{\{(1+\theta)^{m+1} - 1\}(m+2)\theta} - \frac{1}{(1+\theta)^{m+1} - 1}\right) + U_2\right]$$
$$= -\frac{h^3}{12\eta_0}\frac{dp}{dx}\Theta + h\{(U_1 - U_2)M + U_2\}$$

where $\Theta = -12\{\{\quad\}\}$ and $M = (\quad)$.

(b) *Parabolic Temperature Distribution*

$$T = T_1 + t_1\left(1 - \frac{y^2}{h_2^2}\right)$$



$$\eta = \frac{\eta_0}{(1+\theta)^m} \cdot \frac{1}{\left(1 - \frac{\theta}{1+\theta}\frac{y^2}{h_2^2}\right)^m}$$

$$= \frac{\eta_0}{(1+\theta)^m} \cdot \frac{1}{(1 - by^2)}, \text{ where } b = \frac{\theta}{1+\theta} \cdot \frac{1}{h_2^2}$$

also let $\tau = \dfrac{\theta}{1+\theta}$

as before $\dfrac{dp}{dx} = \dfrac{\partial}{\partial y}\left(\eta \dfrac{\partial u}{\partial y}\right)$

whence $\mu = \dfrac{(1-b^2y^2)^{m+1}}{2b(m+1)}\dfrac{dp}{dx} + K_1\int (1-by^2)^m dy + K_2$

K_1 and K_2 integration constants

and $$Q = \int_{h_1}^{h_2} u\,dy$$

$$= \frac{h_1^3}{\eta_0} \cdot \frac{dp}{dx} \cdot \frac{1}{2(m+1)\tau(1-\tau)^m} \frac{1}{(1+\alpha)^3}\left\{-A - \frac{C.D.}{E} + (1+\alpha)B + (1+\alpha)\frac{C.F.}{E}\right\}$$

$$+ h_1\left[\frac{U_1 - U_2}{E} \cdot \left\{F - \frac{D}{1+\alpha}\right\} + U_2\right]$$

where $h_1 = (1+\alpha)h$

$\overset{m}{(1)}$, $\overset{m}{(2)}$. . . . 1st 2nd . . . binomial coeff of order m.

$$A = (1+\alpha) - \overset{m+1}{(1)}\frac{\tau}{3}(1+\alpha^3) + \overset{m+1}{(2)}\frac{\tau^2}{5}(1+\alpha^5) - \overset{m+1}{(3)}\frac{\tau^3}{7}(1+\alpha^7) + \ldots$$

$$B = (1-\tau)^{m+1}$$

$$C = \{(1-\tau\alpha^2)^{m+1} - (1-\tau)^{m+1}\}$$

$$D = \frac{1-\alpha^2}{2} - \overset{m}{(1)}\frac{\tau}{3\cdot4}(1-\alpha^4) + \overset{m}{(2)}\frac{\tau^2}{5\cdot6}(1-\alpha^6) - \overset{m}{(3)}\frac{\tau^3}{7\cdot8}(1-\alpha^8) + \ldots$$

$$E = 1 + \alpha - \overset{m}{(1)}\frac{\tau}{3}(1+\alpha^3) + \overset{m}{(2)}\frac{\tau^2}{5}(1+\alpha^5) - \overset{m}{(3)}\frac{\tau^3}{7}(1+\alpha^7) + \ldots$$

$$F = 1 - \overset{m}{(1)}\frac{\tau}{3} + \overset{m}{(2)}\frac{\tau^2}{5} - \overset{m}{(3)}\frac{\tau^3}{7}.$$

Hence we can once more write :—

$$Q = -\frac{h_1}{12\eta_0}\frac{dp}{dx}\Theta + h\{(V_1 - V_2)M + V_2\}.$$

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Note added in proof.—In the report of the Mechanical Wear Symposium (American Society of Metals, 1950. J. T. Burwell, editor) R. W. Dayton and H. Blok used SAE machines modified to give pure sliding. Both found an appreciable load was carried before scuffing occurred. They apparently assumed no hydrodynamic film could be formed.

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Oilfield Exploration and Exploitation

Geology

1743. A contribution to the study of oil shows. M. Louis. *Rev. Inst. franç. Pétrole*, Jan. 1951, **6** (1), 20–31.—A systematic study of oil shows of various types is of great use in oil prospecting and in tracing the geological processes involved in the origin and migration of oil.

The classification of the various natural bitumens, asphalts, and allied rocks is reviewed; in general, this is based on the different solubilities of the material in organic liquids. Methods of determining the organic materials in rocks and the interpretation of the results of analyses in relation to the C/H ratio are described. C. A. F.

1744. Oil and gas traps. K. K. Landes. *Oil Gas J.*, 8.2.51, **49** (40), 83.—The classification of oil and gas traps is discussed, and a simple classification is presented. Traps can be broadly divided into three groups: structural traps, varying-permeability traps, and combined structural and permeability traps. Structural traps may be subdivided into fold, fault, and fissure traps, and varying-permeability traps into those due to variations in sedimentation, ground water, and truncation and sealing.

Examples of different traps with structure-contour maps and sections are included. These are West Tepetate, Louisiana—anticlinal accumulation; Deep River, Michigan—overlapped dolomite porosity; Apco, Texas—overlapped dolomite wedge; and San Joaquin, Venezuela—dome.

Five references are included. C. A. F.

1745. Types of oil and gas traps in Rocky Mountain region. A. W. McCoy III, R. L. Sielaff, G. R. Downs, N. W. Bass, and J. H. Maxson. *Bull. Amer. Ass. Petrol. Geol.*, 1951, **35** (5), 1000–37.—The part of the Rocky Mountain region discussed herein embraces the states of Montana, Wyoming, Colorado, eastern Utah, and the western parts of North Dakota, South Dakota, and Nebraska. In this area 293 oil and gas traps were discovered up to Jan. 1, 1950. Of these, 177 are in Wyoming, fifty-nine in Montana, forty-three in Colorado, thirteen in Utah, one in Nebraska, and none in North or South Dakota. In these traps an estimated 2,500,000,000 brl of oil and 5 trillion cu. ft. of gas have been discovered.

The 293 discovered oil and gas traps are classified into four major groups: anticlinal, permeability, wedge-out, and fault.

Ten fields illustrating diverse trap types under these major classifications are discussed and depicted by structural maps and cross-sections. The relative commercial importance of each trap type and age of producing reservoirs are compared in a series of maps and bar graphs. Relative importance and trends of discovery methods are also summarized. Conclusions are drawn regarding most productive types of traps and promising methods for their future discovery. E. N. T.

1746. Geology of Erath field, Vermilion Parish, Louisiana. M. H. Steig, I. K. Nichols, S. G. Gray, and R. L. Denham. *Bull. Amer. Ass. Petrol. Geol.*, 1951, **35** (5), 943–87.—The Erath field is in Vermilion Parish in south-central Louisiana, and is of unusual interest, as it is one of the major gas-condensate fields of the Gulf Coast province of Louisiana and Texas. Gas-condensate is produced from numerous sands, and oil accumulation is found in three sands between 7000 and 8000 ft and, in a few places, as a veneer between gas-condensate and water. The main economic importance of this deep dome is due to the fact that the gas-condensate has a high yield of aviation-gasoline and synthetic-rubber constituents. Furthermore, the large, unfaulted, and highly permeable reservoirs form ideal conditions for cycling.

Additional drilling has been done under unit operation, the construction of the cycling plant has been completed, and the cycling operations were started in Apr. 1944. The companies started development of the 7300-ft sands in June 1944; sixteen oil wells were completed by Jan. 1, 1945.

No structural expression of the uplift is present on the surface of the low plain, which ranges in elevation from 6 to 10 ft above sea-level. It is regarded as a deep-seated or non-piercement salt dome.

Gas-condensate saturation exists in nineteen sands of Miocene age ranging in depth from 8100 to 12,000 ft. These reservoirs range in size from an estimated 40 acres to 3000 acres and in volume from 380 acre-ft to 202,000 acre-ft, respectively. Max net sand thickness for individual sands ranges from 7 to 178 ft.

Maps showing structure and net sand thickness of ten of the productive sands, and tables outlining pertinent reservoir data are included. E. N. T.

1747. Several favourable economic factors spur current play in Julesburg Basin. J. A. Kornfeld. *Oil Gas J.*, 11.1.51, **49** (36), 68.—High oil gravities and production, rapid drilling and transport facilities, and multiple reservoirs have encouraged the development of the Huntsman, McLernon, and Dorman fields in Cheyenne County, Nebraska.

480 acres have been proved in the Dorman field, discovered in July 1950. Pays are Dakota D, G, and J sands at approx 4500 to 5000 ft.

In the McLernon field, 4 miles east of Dorman, which was discovered in June 1950, seven wells have been drilled, of which three are producing. The first well initially produced 126 b.d. on pump from the D.

A new shallow pay has recently been found in the Huntsman area, and six gas wells have been completed in the J near Sidney. Oil has been found on the north flank of the Huntsman gas structure.

West of the Huntsman field eight wells have been completed in the Dakota Graneros on a terrace structure.

Drilling and completion practice are briefly described. C. A. F.

1748. Reef limestones of Hermosa formation, San Juan Canyon, Utah. S. A. Wengerd. *Bull. Amer. Ass. Petrol. Geol.*, 1951, **35** (5), 1038–51.—The San Juan country encom-

passes part of the watershed of the San Juan River in Arizona, Colorado, New Mexico, and Utah. It covers the north-central part of the Colorado Plateau province, and shows great diversity of structure and stratigraphy.

The Hermosa formation, exposed in the San Juan Canyon, where it transects the Monument upwarp in southeastern Utah, contains several biostromal layers, one of which changes laterally into small bioherms with typical reef-limestone breccia making up the greater part of the deposit.

A bioherm excellently exposed in the San Juan Canyon walls was examined just above river level, between Mendenhall loop and Honaker trail. At least eight bioherms have been breached by the San Juan River, several of which are exposed on only one canyon wall. The reef limestones are present through approx two-thirds of the Hermosa thickness below the Hermosa–Rico gradational contact, and the fossils suggest that the bioherms are Des Moines in age.

Regionally, the biohermal trends may be controlled by a shelf related to the Paradox–Salt Valley basin southeastern Utah and southwestern Colorado. Valid predictions of possible distribution of these reefing limestones must be based on further surface examination, subsurface analysis, and an appraisal of regional controls exerted by the Paradox evaporite basin in relation to the Colorado Mountain highlands.

Experience with both recent and ancient reef limestones suggests that bioherms should not be considered abnormalities in the proper stratigraphic section. All sedimentary rocks are products of their many controlling environments, and it behoves the exploration scientist to deduce optimum conditions of neritic slope, depth of water, type of bottom, and supply of calcium carbonate in relation to clastic sediment distribution for the max development of bioherms.

The Hermosa bioherms of the San Juan Canyon area are small knob-like masses of reef limestone found in the lower Hermosa section. They are small where now known, and certain apparently correlative, thick, gas-producing limestones suggestive of reef limestones in the Barker Dome field, in the northwestern San Juan Basin, make it possible that a long arcuate trend of Des Moines reefs will eventually be delineated by exploration and drilling in southeastern Utah. E. N. T.

1749. McIntosh formation, Centralia-Chehalis coal district, Washington. P. D. Snavely, Jr., W. W. Rau, L. Hoover, Jr., and A. E. Roberts. *Bull. Amer. Ass. Petrol. Geol.*, 1951, **35** (5), 1052–61.—The name McIntosh formation is proposed for more than 4000 to 4500 ft of dark-grey marine siltstone and claystone, and interbedded arkosic and basaltic sandstone which crops out in the Centralia–Chehalis coal district, Lewis and Thurston counties, Washington. The siltstone and claystone beds contain a fauna correlative with the Laiming's B–1A foraminiferal zone of upper Domengine age of California. The McIntosh formation is correlated with the Mill Creek and Sacchi Beach beds of local usage and perhaps with the Burpee formation of Oregon.

The McIntosh formation consists chiefly of dark-grey well-indurated tuffaceous siltstone and claystone with thin interbedded tuff zones. Some beds are massive, although stratification may be made apparent by tuffaceous zones or interbedded sandstone. Irregularly located calcareous nodules and lenses are common. The siltstone and claystone are fissile in part, and are commonly finely micaceous. Carbonaceous material and pyrite are present in most places. Most beds are laminated, consisting of fine layers of siltstone and fine-grained tuff or sandstone. The sedimentary rocks weather into an iron-stained shaly and crumbly soil, the colour of which, in most places, is a mottled light grey and light yellowish orange. E. N. T.

1750. Faults in sedimentary part of Wichita Mountains of Oklahoma. B. H. Harlton. *Bull. Amer. Ass. Petrol. Geol.*, 1951, **35** (5), 988–99.—In a field study in 1936–37 along the flanks of the Wichita Mountains in Oklahoma, the large area of Cambro–Ordovician rocks in the northeast part of this mountain complex was mapped in detail. Several structural features were disclosed which were at variance with published reports by Taff and Becker and with information shown on the geologic map of Oklahoma.

By careful examination of the exposures, supported by information from air photos and other topographic data, it was possible to subdivide the sedimentary area into two main structural blocks, both of which contain subsidiary features almost as prominent as the blocks themselves. The purpose of this paper is to record the observations which reveal the relationships of these units and clarify the structural features within them.

The Cambro–Ordovician sediments were found in two structural blocks: a graben adjacent to, and sub-parallel with, the igneous core of the Wichita Mountains, and a horst bordering the graben on the northeast and serving as the limiting structural feature of the mountain complex. These structural blocks are sufficiently important to be termed the Lawtonka graben and the Blue Creek horst, respectively. E. N. T.

1751. Rumblings from Manitoba. P. C. Ingalls. *Oil Gas J.*, 8.2.51, **49** (40), 132.—15–18 Daly, approx 600 miles to the east and south of the Alberta fields, has indicated oil in commercial quantities. The well is in the shallow northeastern shelf area of the Williston Basin, and was sited after intensive geological and geophysical work in the area. Pay is believed to be Devonian. C. A. F.

1752. Some new ostracods from the upper Senonian of Cameroun. N. Grekoff. *Rev. Inst. franç, Pétrole*, Feb. 1951, **6** (2), 53–9.—Three new species and one new genus of ostracoda from Cameroun are described. One of these, *Buntonia howe*, previously regarded as restricted to the Cainozoic, has been found in the Upper Senonian. Line drawings of the fossils are included. C. A. F.

1753. Robulus incisus n. sp.; a characteristic of the Palæocene. M. Lys. *Rev. Inst. franç. Pétrole*, Apr. 1951, **6** (4), 91–100.—It has been found that the fossil species *Robulus incisus* n. sp., which occurs in the Tertiary Senegal basin, is characteristic of the Palæocene formations. It has also been found in the Palæocene of Morocco and the Lower Eocene of Algeria. C. A. F.

1754. The Georgsdorf field. H. Lötgers. *Erdöl u. Kohle*, 1949, **2** (12), 537–44.—Development of the Georgsdorf (Emsland) field began in 1943. Since 1947 numerous new wells and increased production have greatly increased knowledge of the geological structure and reservoir conditions, but the description of the stratigraphy published then is still valid.

The productive area is limited in the east by an increase in clay content and thinning of the reservoir rock. Edge water has been found both in the north and south, but the western limits have not yet been reached.

Although the general picture of the Georgsdorf fold dominated by axial faults still holds, the details of the structure are now found to be much more complicated, and the author divides the field into five structural units which he describes in some detail. The tectonics of West Emsland are those of a series of broad, east–west folds caused by movements at various periods, commencing in the Lower Kimmeridge. Georgsdorf is only part of this regional structure, but the intensity of the folding, especially in the eastern part of the field, does not accord with the otherwise gentle structural forms, and it is suggested that additional uplift may have been caused by deep-seated salt in the Münder Mergeln beds of the Portland. S. E. C.

1755. Petroleum geology of the Swiss Molasse basin. H. M. Schuppli. *Erdöl u. Kohle*, 1950, **3** (8), 361–8.—Since Heim's work in 1919, knowledge of the petroleum geology of the Swiss Molasse basin has greatly increased, and details of investigations and results are now being published officially.

The possibility of oil in Switzerland is suggested by shows, of which there are four kinds: asphaltic seepages and impregnated rocks in the Jura, oil sands in the Molasse of the sub-Jura zone of central Switzerland, gas shows in the sub-Alpine zone, and light oil shows in the Rupelian Beds of the sub-Alpine Molasse of the upper Entlebuch. These are discussed in detail. Structural conditions in Central and West Switzerland are not unfavourable for migration and accumulation, but some knowledge of the deeper structure, such as would be given by seismic profiles, is necessary before a definite opinion can be expressed. Sedimentation was such that stratigraphical traps are also a possibility. Some particulars are given of previous boreholes, all of which failed because of unfavourable locations and bad handling. It is suggested that more drilling should be undertaken, and for stratigraphical and structural reasons it should start just south of the sub-Jura zone. S. E. C.

Drilling

1756. Deep rotary drilling. Anon. *Oil Gas J.*, 5.10.50, **49** (22), 141.—By means of photographs, illustrations, and captions complete drilling operations from location and rig erection to cementing casing are described. C. G. W.

1757. Fluid drives for drilling rigs. K. N. Mills. *World Oil*, Jan. 1951, **132** (1), 87.—Of the two types of fluid drives, the greater flexibility of the hydrokinetic drive, utilizing fluid velocity, has found wider application than the positive displacement or hydrostatic drive. The characteristics of hydraulic couplings and torque converters are considered. One advantage of hydraulic couplings is that the engine may start at no load and attain a higher speed before taking the load; this is of particular significance in the acceleration of dead loads. Comparison of fluid-drive and friction-clutch performance curves emphasizes the advantage of the former. The torque converter ensures that the engine is not subjected to severe overloads, and finds suitable application also with slush pumps. A. J. H.

1758. New portable drilling mast. R. F. Carlson. *Oil Gas J.*, 5.10.50, **49** (22), 190.—A new design of mast affords more visibility and floor space than conventional forms. The mast is in the form of an A-frame joined at the crown and pivoted at the base. Auxiliary legs assist in raising the frame to the vertical position, where it is securely locked. The legs of the A-frame are of latticed-box construction. The gross capacity of the mast is 566,000 lb, height 133 ft. C. G. W.

1759. Portable folding masts for drilling and well service. A. Vierling. *Erdöl u. Kohle*, 1951, **4** (2), 66–75.—The various types of mast and derrick for portable drilling and servicing equipment are described, and the main factors in design indicated. The types are divided into three groups: for shallow exploration work, for medium and deep drilling, and for servicing producing wells. Important points in care and maintenance are noted. There has been a rapid development of this equipment in America during and since the war, but there are now available satisfactory German designs. S. E. C.

1760. Tour of the oilfields of the U.S.A. F. Brüggemann. *Erdöl u. Kohle*, 1951, **4** (3), 109–15.—A brief account of outstanding features in drilling and production practice seen during a six weeks' tour of the American fields. S. E. C.

1761. Buckling of rotary drilling strings. Pt I. A. Lubinski. *World Oil*, Mar. 1951, **132** (4), 105.—The subject is approached mathematically, but derivation of formulæ is confined to an appendix.

In a straight hole, the drill string remains straight, so long as the bit weight is below the critical value, and any increase in weight is thereafter accompanied by buckling of the first, second order, etc. The effect of drill collars is to increase the value of the critical weight; thus drill strings with no drill collars buckle easily. The heavier the mud the smaller the critical weight. The rotary percussion method is known to give straighter holes. Fatigue failures are due to alternation of stress in a buckled string. Bending moments and stresses are considered.

The force F on the well wall increases to a max at buckling condition, and then falls rapidly. Thus it is preferable to maintain conditions above the critical, rather than just below. F is not large, but, as a function of apparent hole radius, any caving occurring in the well must be accompanied by an increase.

Extensive data are given in twelve graphs and references. A. J. H.

1762. Methods of calculation of the length of a multilayer rope winding on a winding drum. W. Tyminski. *Nafta*, Jan.–Feb. 1950, **6**, 8–10.—Formula for length of rope on drum L when i layers of rope of dia d are wound on a drum of dia D and width E is:

$$L = \frac{i\pi E}{d}\left[D + d + \frac{\sqrt{3}}{2}d(i-1)\right].$$

This is proved and an example is worked out. M. S.

1763. Causes of frozen drill pipe and casing. M. Gawlinski. *Nafta*, Mar. 1950, **6**, 50–5.—Author summarizes laboratory and field experiments carried out to investigate the causes of frozen drill pipe and casing and suggests remedies to be applied in such cases. M. S.

1764. Fatigue testing and development of drill pipe to tool joint connexions. W. S. Bachman. *World Oil*, Jan. 1951, **132** (1), 104.—Analysis is given of information collected over a period of fifteen years involving more than a thousand tests made by the Hughes Tool Co. laboratory. After a brief introduction to the development of rotary tool joints, the factors contributing to failure are considered separately, and specific cases quoted. The survey covers the various types of pipe connexions.
References. A. J. H.

1765. Effect of mud pump surges. H. C. Wachtman. *World Oil*, Jan. 1951, **132** (1), 94.—Previous work on slip velocity of falling particles was confined to smooth flow conditions. The experiments described were conducted to determine the effect of certain variables on the carrying capacity of muds: weight and shape of particles, velocity and visc of muds, and amplitude and frequency of surges. Observations were made on the net velocity rise of particles in a 2-inch-dia vertical plastic tube. The laboratory colloidal suspensions were made up of bentonitic material. It is concluded that surges tend to increase the slip velocity.
Experimental procedure and results are given. References. A. J. H.

1766. Electric log resistivity, porosity, and water saturation for clean and shaly sandstones. P. J. Jones. *World Oil*, Feb. 1951, **132** (2), 140.—The use of Archie's equation for determining the porosity and water saturation from electric log data had been restricted to clean sandstones. Consideration is given to the effects of shale which may exist as a cementing material, as a shale bed, or scattered within the pores of the sand. Allowing for certain assumptions, an equation is proposed for use with electric logs, and an explanation given for the difference between electric log data, uncorrected for shale content, and practically determined information relating to oil content of shaly reservoirs.
References and graphs. A. J. H.

1767. Loss of circulation of drilling mud in wells. M. Gawlinski. *Nafta*, Jan.–Feb. 1950, **6**, 1–8.—Author discusses causes, remedies, and methods of prevention of loss of circulation of mud. Causes are: (1) high pressure; (2) low pressure; (3) cracks in rocks. To stop loss of circulation caused by (1) gels are recommended (preferably inorganic) or lowering of mud density or cementing. Knowledge and experience of particular kind of rock worked would enable the driller to suggest preventives. To counteract losses due to (2), bentonite addition is recommended or cementing, which is found particularly effective when 6–8% of bentonite is added to cement. Any sudden loss of circulation can be assumed to be caused by (3). Prompt cementing or insertion of casing is the only remedy. M. S.

1768. Some controlling factors regarding variable weighting of cement slurries. M. A. Mallinger. *Petrol. Tech.*, Dec. 1950, **11** (12), 10.—In order to avoid loss of returns in cementing operations where lost circulation is a problem and where high hydraulic pressure must be avoided, weight-reducing additives may be desirable in cements. Study has shown that bentonite is a satisfactory additive. 20% of bentonite (relative to cement-weight) gave reductions from 16·2 to 13·1 lb/gal in one case, and from 14·2 to 12 lb/gal in another. The max allowable bentonite content for a 100 p.s.i. tensile strength at 24 hr was determined at temp from 80° to 200° F, and ranged 3 to 16%. Usable amounts of bentonite did not appreciably change pumping time if the initial consistency of the slurry with bentonite was the same as for the slurry without bentonite. Bentonite increased the permeability and porosity of the set cement. Even with the max amount of bentonite the air permeability was less than 1 mD. Porosities attained 55%.
The effectiveness of bentonite is apparently associated with its water-absorbing properties, and the cement has a higher water content than in the absence of the additive.

There is a limited demand for heavy cements. Barytes has been found to be a suitable additive provided that the cement is carefully selected. In some cements the barytes adversely affects the cement-setting characteristics. The pumping times were little affected, and the same was true of the permeability. There was some effect on porosity. Weights of 19 lb/gal have been obtained. G. D. H.

1769. Recovery of weighting material from muds. W. Gründer. *Erdöl u. Kohle*, 1950, **3** (9), 442–5.—There are few processes for the separation of fine-grained weighting material from mud, but its value is sufficient to make any new method of interest. The process described was developed for special cases of ore treatment, but it gave good results when tried on a small scale with heavy muds. It is based on an observation during flotation experiments, when it was seen that heavy minerals accumulated in a ring immediately below holes in the periphery of a bell-shaped stirrer. A cylindrical stirrer with slits and grooves was therefore made so that it could be lifted, while rotating, in and out of the mud. On lowering into the mud the grooves quickly fill with heavy minerals, and in 20 to 30 sec it is lifted, the grooves then being swept clean by a brush which pushes the minerals on to a collecting tray. It was found that with suitable mud densities 80 to 90% of the heavy minerals could be removed. The building of a plant for handling used mud is discussed. S. E. C.

1770. Horizontal centrifuge desands contaminated mud. A. Gibbon. *World Oil*, Mar. 1951, **132** (4), 99.—Abrasion due to sand particles in drilling is a danger to equipment and drilling procedure and should be prevented. The horizontal centrifuge has an eff of 84·7%, and is driven at 1200 to 1600 r.p.m. by a 130 max h.p. motor. Because of its high eff and rate of throughput of 500 gal desanded mud per min, the unit need not be operated continuously, but only for such time as the mud contains much sand. It is claimed that sand content of mud in an average well may be reduced from 15 to 1% in 4 hr. A. J. H.

1771. The rheology of suspensions. Application to drilling muds. Pt 2. The elastic properties of muds. R. Métrot. *Rev. Inst. franç. Pétrole*, Feb. 1951, **6** (2), 39–52.—Methods are described of investigating the elastic properties of clays, in which the material under test is subjected to deformation under known tangential stresses. Three methods are available for investigating these properties; these are: the capillary tube method, in which a cyl of the material is deformed under a differential pressure between the ends of the tube; a method in which the sample is placed in a vessel containing a thin metal plate suspended vertically from a balance-pan whose weight can be adjusted, the movement of the plate being observed optically; and the coaxial-cyl method, in which the sample is contained in the annular space between two coaxial cyls, the outer one of which is fixed and the inner suspended on a torsion wire. The wire is turned through a given angle, and the corresponding rotation of the inner cyl is observed, the angular difference being a measure of the elasticity of the sample.

The elastic properties of muds are characterized by five constants: the moduli of rigidity, the reactivity, the yield value, and the visc.

The application of a knowledge of these properties in the use of drilling muds is outlined. C. A. F.

1772. The rheology of suspensions. Application to drilling mud. Pt 3. The yield value of drilling muds. R. Métrot. *Rev. Inst. franç. Pétrole*, Mar. 1951, **6** (3), 67–82.—The most important of the rheological properties of drilling muds is the "yield value," which can be defined as the tangential force which must be applied to the material to convert it from the solid to the liquid state.

Three methods for the determination of the "yield value" are described in detail. These are: the capillary tube method, the shearometer method, and the coaxial-cyl method.

Very dilute clays have no plasticity and no "yield value." As the concentration is increased the "yield value" becomes measurable at the "liquid limit." Above this, with increasing concentration, the "yield value" increases rapidly, and at the "plastic limit" approaches infinity.

Thixotropic change and the metaplastic states are discussed. C. A. F.

Production

1773. Wave forces and vertical loading on offshore well casing. J. A. Reed and H. H. Meredith. *World Oil*, Jan. 1951, **132** (1), 137.—In the design of well-heads in offshore locations, account must be taken of the unstable casing columns. A typical design for a casing programme is illustrated, and the procedure described. The forces developed by breaking waves are estimated by reference to Munk's theory, based on the Solitary Wave and Airy Wave theories, and on the data available on wave heights. Neither of these basic theories take into account breaking waves, which give the max force, and it was decided, therefore, to consider only those values which correspond to the worse conditions. Graphical analyses of load diagrams are explained.

References and diagrams. A. J. H.

1774. Influence of production practice from natural gasoline recovery from condensate fields. B. H. Sage. *Oil Gas J.*, 1.2.51, **49** (39), 65.—A typical condensate-type reservoir is considered and evaluated. The gas phase assumed at the retrograde dew point is contained in a consolidated sand wet with water. Reservoir pressure is 4630 p.s.i.a. and temperature 250°. Permeability porosity and sand thickness are assumed uniform throughout the reservoir. Two types of production operations are considered: (*a*) withdrawal from several points in the reservoir; (*b*) gas-cycling operation using propane free gas.

Calculations are performed assuming equilibrium conditions obtained. It is estimated in this hypothetical case that if gas cycling had not been carried out approx 15% of propane and butanes and 37% of pentanes and heavier hydrocarbons would have remained underground when field pressure had decreased to 10% of its original value. C. G. W.

1775. Retrograde condensation and condensate fields. C. Schmid. *Erdöl. u. Kohle*, 1950, **3** (9), 422–7.—The phase relationships of the lighter hydrocarbons are described and the phenomenon of retrograde condensation explained. There are numerous reservoirs in the U.S.A. in which the hydrocarbon content, although very mixed, was originally in a single gaseous phase but from which, on expansion, a liquid phase separates. Examples are given, and it is pointed out that although no such reservoirs have yet been discovered in Germany they may well be there at great depth and if retrograde condensation is not taken into account when they are discovered, production losses will ensue. S. E. C.

1776. A study of the Dornick Hills–Springer sand reservoir, Velma pool, Oklahoma. W. B. Davis. *Petrol. Tech.*, Jan. 1951, **3** (1), *A.I.M.M.E. Tech. Paper No.* 2999, 29–36.—The reservoir is a series of sand bodies in the Pennsylvanian on the northeast flank of the Velma anticline and dips northeast at about 45°, but there is an easterly trend in the south, where the dip averages 25°. There is faulting. The up-dip productive limit is fixed by truncation; the down-dip limit is due to a gradual reduction in sand permeability. Most wells found two substantial sands, but some found deeper sands also.

In 1944 a well found Dornick Hills–Springer production; by June 1950 there were 274 oil wells and six gas wells, and these constituted almost full development except at the southeast end. Some wells flowed initially, but most pumped 200 to 500 b.d., though many could have pumped 1000 to 3000 b.d. if pumped down.

At −4500 ft the original pressure was 1670 p.s.i.g. Tests on bottom-hole samples at various depths indicate gas saturations appropriate to the structural position. The sand is generally fine-grained with argillaceous material. The average interstitial water content was 20%; average porosity is 18%, and permeability 150 mD, with the oil permeability when water is present 50 to 70% of the dry air permeability. The average formation volume factor is 1·19. Oil in place in the first and second sands is estimated at 430,000,000 brl.

There is no significant water influx to date, and the effective producing mechanisms are expected to be dissolved gas and gravity drainage. 29,000,000 brl of oil had been produced to June 1, 1950, the average pressure drop being 59,000 brl/p.s.i. The general decrease in G.O.R.'s over the period is thought to indicate the effects of gravity

drainage. Lamination in parts of the reservoirs may reduce the efficiency of recovery by gravity drainage.

Solution-gas drive should give 18% recovery, and gravity drainage may raise the ultimate recovery to 37%, although much of the gravity drainage oil may be recovered over a long period at low rates. G. D. H.

1777. Performance of the Lower Pawelek reservoir, Falls City field, Karnes County, Texas. J. W. Crutchfield and E. F. Bowers. *Petrol. Tech.*, Dec. 1950, **2** (12), *A.I.M.M.E. Tech. Paper No.* 2937, 335–44.—The Falls City field is on the southern downthrown side of a fault, and has twelve reservoirs at 4600 to 6050 ft. Sixty-four wells have been completed, thirty-one being in the Lower Pawelek sand, which is 25 to 40 ft thick. There was no gas cap, and the oil column was 53 ft thick. The sand averages 24·5% porosity, with permeabilities of 311 mD parallel to the bedding and 121 mD perpendicular to it. The connate water is estimated at 37%. The structure is a flat dome, and the average net oil-sand thickness is 16 ft over 919 acres. The average well density is 34 acres per well. Cumulative production at the beginning of March 1949 was 694,281 brl, and some low wells were averaging 20% of water. The pressure had dropped from 2625 to 2281 p.s.i. Five wells were on artificial lift, and tubing pressures in the others were low and critical as regards maintaining natural flow.

Little gas was available locally for gas lift, water disposal was a problem, and the installation of conventional pumps seemed inadvisable. Calculation showed that the injection of 1500 b.d. of water and a production of 2000 b.d. would maintain a pressure of 2400 p.s.i.

A closed salt-water-gathering system was erected, and water was injected into a dry hole between the edgewater level and the fault and midway along the extent of the field, *i.e.*, at a point most remote from natural water drive. Injection pressures ranged 400 to 600 p.s.i., and rates rose from 500 to 1800 b.d. in June 1948. After injecting 100,000 brl of water the pressure had risen 64 p.s.i., although 213,000 brl of reservoir fluids had been withdrawn in the same time. A net withdrawal rate of 500 b.d. was maintained, and in April 1950 the pressure was 2369 p.s.i. The downward trend in tubing pressures was arrested, and in some cases the pressure has risen 100 p.s.i.

Pressure and structure maps, a cross-section, production-data curves, and other data are presented. G. D. H.

1778. Reservoir analysis of the Redwater pool. I. Haskett. *Canad. Min. Metall. Bull.*, Apr. 1951, **44** (468), 226–36.—The history of development, regional geology, reservoir characteristics, and pressure-production history are described. Production is from a Devonian coral limestone reef at an average depth of 3150 ft. The estimated productive area of the pool is about 37,000 acres, the oil zone having a max thickness of just over 200 ft. There are wide variations in both type and values of porosity, the average porosity being 6·16%. Total producible oil is about 1·24 billion brl. The crude oil has a saturation pressure of 485 p.s.i. compared with the original reservoir pressure of 1,050 p.s.i., and the principal recovery mechanism has been, so far, a natural water drive. Cumulative production to Jan. 1951 has been about 15½ million brl, and has resulted in a decreased reservoir pressure of just over 100 p.s.i. Predictions indicate that the natural water drive will be insufficient to maintain the reservoir pressure above saturation pressure after 1953 and that, with the natural depletion of the pool, production will be mainly by a solution-gas drive. W. H. C.

1779. Applications and results of the Hydrafac process. W. E. Hassebroek. *World Oil*, Feb. 1951, **132** (2), 133.—This process is more suited to the treatment of wells which exhibit an abnormal permeability or a sudden decrease in production. The principle of operation consists in subjecting the formation to a high pressure, in excess of the overburden pressure, by the injection of a high-visc gel which penetrates the sand at a low rate. Sand grains of approx 0·026 inch dia within the gel assist in maintaining the fractures in the sand body. At the termination of the operation, the gel is broken down and production re-commenced.

Several cases are taken as examples of the eff of the process, and the results of tests and local procedures are discussed. The recent introduction of a combination Hydrafac–acid-treating system is mentioned. The process is also used as a well-completion programme. A. J. H.

1780. Directional permeability and dimensional orientation in Bradford sand. J. C. Griffiths. *Producers' Monthly*, June 1950, **14** (8), 26.—Two important properties of sedimentary rocks are their mineral composition and textural characteristics, the latter including orientation. In petrofabrics, the study of orientation in rocks, two types of orientation are investigated: dimensional, or grain, and lattice, or structural orientation. It was decided to consider dimensional orientation in sedimentary rocks; three Bradford cores, the permeability trends of which were known, were selected and fourteen thin sections taken at various angles. The sections were projected on to paper and permanent records taken of: (*a*) elogation, the ratio of the dia of min circumscribing circle to that of max inscribing circle, and (*b*) inclination of grain axis. Difficulties were encountered in expressing the results graphically, but it was found convenient to plot frequency diagrams of number of grains versus the inclination from the reference direction.

Variations in permeability have been generally associated with the planes parallel to, and perpendicular to, the bedding planes. Of the section taken perpendicular to the bedding plane, definite orientation was noted along the max permeability direction, and to a lesser extent along the direction of min permeability. The orientation in the sections parallel to the bedding plane were inconsistent.

The results of the tests can only be used to indicate certain trends, and are not meant to be exhaustive. The observations are correlated with the sedimentation characteristics of the sand. The transportation behaviour of pebbles and of sand grains is compared.

References. A. J. H.

1781. Pressure behaviour in the Woodbine sand. J. S. Bell and J. M. Shepherd. *Petrol. Tech.*, Jan. 1951, **3** (1), *A.I.M.M.E. Tech. Paper No.* 3000, 19–28.—Forty-two fields have produced from the Woodbine sand, and twenty-eight fields now produce, the 1949 output being 118,000,000 brl. Except in one case the drive has been predominantly by water expansion. Now pressure data have been obtained in non-productive areas. The Woodbine consists of thick blanket sands with interbedded shales. Oil production is from domes, fault traps, and pinch-outs. In the East Texas Basin the Woodbine covers 31,500 sq. miles. Two main areas produce—the Mexia–Talco fault line and East Texas.

It was recognized that pressure in this common aquifer would decline with production.

Shortly after finding oil in the Woodbine at Hawkins in 1940 it was noted that a major fault divided the field into two parts; in the eastern part the original pressure at −4075 ft was determined as 1710 p.s.i., and in the western part 1990 p.s.i. The gas/oil and oil/water contacts in East Hawkins were considerably lower than in West Hawkins, and the pressure in the latter has been concluded to be normal for the depth in the Woodbine. It is inferred that West Hawkins is a closed reservoir, separated by an impermeable barrier. East Hawkins was 280 p.s.i. below normal, probably due to withdrawals at East Texas over ten years.

Original Woodbine pressures have been estimated for various depths, and these have been used to derive pressure losses at the various wells at which pressures have been determined. The pressure losses have been contoured on a map, and the contours indicate that East Texas has been the main cause of pressure drop, but the fault-line fields have also contributed significantly. Plots have been made for pressure drop against the logarithms of the distance from East Texas, and there was a general straight-line trend. The effects of East Texas extend outwards for 65 miles. A similar plot has been made for the area affected by fault-line production, and the effects extend some 80 miles. Certain pressure-loss-contour peculiarities lie in the zone which is in the radius of influence of both East Texas and the fault-line fields.

Some pressure irregularities arise from fault interference. G. D. H.

1782. Capillary pressure method of interstitial water determination. W. Rose. *Oil Gas J.*, 5.10.50, **49** (22), 212.—It has long been appreciated that connate water, irrecoverable oil, and approx rock-texture values could be estimated from capillary pressure data. Further, suitable laboratory core studies could improve electric-log interpretations. The difficulties of planning and performing experiments on cores and of interpreting the results are fully discussed. The need for the application of data on

accumulation and recovery processes and for statistical interpretations of results is emphasized. This approach requires the attention of geologists as well as petroleum engineers.

References are appended. C. G. W.

1783. Elements of dynamometer tests. D. S. Stear. *World Oil*, Feb. 1951, **132** (2), 160.—Three main types of dynamometers are in general use: hydraulic, mechanical, and electrical. Recently a steel-ring dynamometer of the mechanical type has been produced, and besides giving good performance, is simple and easily installed. Dynamometer tests are, as yet, the only positive source of information for designing or checking well conditions, but various empirical formulæ have been developed for the calculation of well loads. Of these, the Slonneger (API), Mills, Langer, and Caraway formulæ are given, and their range of application reported. Thus the Slonneger formula is suitable for pumping speeds up to 20 strokes per min, the Mills up to 25 strokes per min, and above these speeds, Caraway has presented his own; this is similar to the Mills formula.

References. A. J. H.

1784. Calibration characteristics of subsurface gauges. R. V. Smith, E. J. Dewess, and R. H. Williams. *Petrol. Engr*, Jan. 1951, **23** (1), B-7.—In order to convert the deflection of a pressure-sensitive element into pressure units it is necessary to know the gauge modulus, the zero pressure correction, and the curvature correction of the gauge. The gauge modulus and zero pressure correction may be determined by applying the method of squares to the data obtained in calibrating the gauge over a range of temps and pressures. Curvature corrections are defined as deviations from a linear pressure–deflection relation. Average deviations for a number of observations at each pressure are determined.

Pressures are then determined by a relation

$$P = mD + a + c$$

where P = pressure applied, p.s.i.;
m = gauge modulus, p.s.i. (pressure corresponding to a change of deflection of 1 inch);
D = deflection, inch (pressure deflection of the stylus from the zero-pressure line);
a = zero-pressure correction, p.s.i.;
c = curvature correction, p.s.i.

Although curvature corrections could be made by fitting observed calibration data to a higher-order relation, the linear method is simpler and more convenient.

The immediate history of a pressure gauge may also affect the pressure-deflection relation. If the pressure on a gauge is increased in small increments and then decreased, the deflections produced by the same pressures will be greater when the pressure is falling than when the pressure is rising. This effect is due to hysteresis. This effect can only be corrected by a special calibration or by taking care to avoid conditions producing hysteresis effects.

The application of these corrections to deflections of subsurface pressure gauges is described. References are appended. C. G. W.

1785. Electrical bottom-hole pressure gauge. C. H. Biedendieck. *Erdöl u. Kohle*, 1950, **3** (8), 383–4.—A stretched steel wire has one end attached to a diaphragm so that variations in borehole pressure alter the tension in the wire. Damped vibrations are induced in the wire by mechanical or electrical means, the frequency varying with the tension. The electrical counterpart of these vibrations is produced at the surface on a cathode-ray tube, and superimposed on them are the effects from a similar vibrating wire fitted in the surface equipment. The tension in this second wire can be adjusted until both vibrate at the same frequency, and if the tension-regulating device is suitably calibrated a direct measurement of bottom-hole pressure is obtained. The range of the instrument can be extended to 250 atm. S. E. C.

1786. Electrolytic model studies as applied to water-flooding a shoestring sand. W. E. Eckard and J. A. Mason. *U.S. Bur. Mines Rep. Invest. No.* 4778, Mar. 1951.—An electrolytic model investigation is described of the well patterns and spacings required economically to water flood the Paola shoestring field in Kansas.

The field is 8 miles long and varies from 200 to 700 ft in width. The pay is Pennsylvanian Cherokee Squirrel sand, which has a max thickness of 60 ft along the axis of the shoestring and wedges out on either side.

The electrolytic equipment used was similar to Botset's model. The shape of the electrolytic model field, which is made of gelatin, is designed to give a cross-sectional area comparable to an average cross-section of the pay of the shoestring. The principle of the model is to transfer positive copper ammonium ions in a copper ammonium chloride solution from the positive terminal of direct current to the negative terminal through a gelatin field containing zinc ammonium chloride ions. Blue copper ammonium ions moving through the field displace colourless zinc ammonium ions and form a pattern conforming to the volume of the reservoir that would be flooded by injected water.

A number of different well arrays were used, and it was found that the most efficient flooding technique was given by a two-stage pattern, the first having alternate rows of two injection wells and two producing wells across the width of the shoestring, and the second, a modified " seven-spot " pattern. C. A. F.

1787. Correlation of bottom hole sample data. G. Borden and M. J. Rzasa. *Petrol. Tech.*, Dec. 1950, **2** (12), *A.I.M.M.E. Tech. Paper No.* 2931, 345–8.—Charts are given for bubble-point pressure correlation and for reservoir volume-factor correlation. Published data for three samples of oil are used to test the correlations. Tables compare predicted and experimentally determined bubble-point pressures and reservoir-volume factors. G. D. H.

1788. The effect of permeability stratification in complete water-drive systems. M. Muskat. *Petrol. Tech.*, Dec. 1950, **2** (12), *A.I.M.M.E. Tech. Paper No.* 2894, 349–58.—General equations are given for calculating the performance of complete water-drive reservoirs producing from idealized stratified formations in which the permeability stratification is either exponential or linear. Different degrees of permeability stratification are considered, and special emphasis is given to the effect of the mobility ratio between the produced oil and invading water. Curves show the initial break-through recovery as a function of mobility ratio for different degrees of permeability stratification, and the composition of the fluid output as a function of cumulative oil recovery.

It appears that when the mobility of the oil is less than that of the invading water the channelling tendency due to permeability stratification is aggravated as the higher permeability zones are flooded out. On the other hand, when the oil mobility is high compared with that of the invading water, the flooding of the high-permeability zones will have a retarding and choking effect, thereby partially suppressing the gross by-passing phenomena.

The various basic assumptions are discussed, including the influence of ignoring the stripping phase. G. D. H.

1789. Oil displacement calculations. J. W. LaRue and D. A. Wilson. *World Oil*, Jan. 1951, **132** (1), 146.—The effect of relative-permeability data on certain recoveries is investigated with reference to four sands. The displacement history was calculated on a basis of visc ratio; the cores were assumed to have a connate water saturation equivalent to that for zero water flow, and the remainder of the pore space was taken to be saturated originally with gas-free oil. The method of calculation is explained with reference to graphs. It is concluded that the relative-permeability characteristics of a core have little effect on water-flood eff.

References. A. J. H.

1790. Open flow potential calculation for dual completion wells. C. W. Binckley and W. L. Hixon. *World Oil*, Feb. 1951, **132** (2), 150.—The completion of a well running two tubings, 2⅜- and 1-inch O.D. within a 5½-inch casing required the development of an equation for the equivalent dia of cross-section; previously there existed equations only for single uniform annular flow. The underlying theory is given with particular reference to field data, and the well completion is explained. References. A. J. H.

1791. An investigation of the electrokinetic component of the self potential curve. M. R. J. Wyllie. *Petrol. Tech.*, Jan. 1951, **3** (1), *A.I.M.M.E. Tech. Paper No.* 2940, 1–18.—A laboratory investigation of the relationship between the streaming potential

developed across a mud filter-cake and the pressure differential causing filtrate flow has been carried out, using eight water-base muds prepared in the laboratory and fifteen field-mud samples. It was found that $E_s = kP^y$, where E_s is streaming potential, P the pressure differential, and k and y constants for a given mud at a constant temp.

Beyond a minimum cake thickness the relationship is independent of this factor, while for some systems there is a value of P above which the above simple relationship ceases to hold. k tends to be dependent on the mud resistivity at 77° F, but y has not shown any relationship with any commonly measured mud property.

Field experiments have shown that the total S.P. (self potential) is the algebraic sum of an electrokinetic and an electrochemical potential. A chart permits the use of mean solution activities in interpreting electrochemical S.P. data.

The use of low-resistivity drilling muds is essential if optimum accuracy is desired in using the S.P. curve to calculate connate-water salinities by the electrochemical S.P. method. G. D. H.

1792. Mounting core analysis specimens in thermoplastic. W. E. Showalter. *Petrol. Tech.*, Dec. 1950, **11** (12), 8–9.—The consolidated specimen is shaped and cemented into the lucite container, which has been machined to a close fit. The hermetically sealed plastic-enclosed specimen is placed in a pressure bath, and the pressure is raised to about 500 p.s.i. and the temp to 250° F. The softened plastic is compressed against the rock and penetrates the outer layer. The bath is cooled under pressure after 4 hr, and then the mount machined and polished as required. There is good bonding and penetration of about half a grain dia. If needed, the ends can be protected by blotting-paper discs, so that they can later be bared without cutting the specimen.

Unconsolidated matter can be mounted similarly, but some lateral support is needed during the softening process in the bath. G. D. H.

Oilfield Development

1793. Oil and gas developments in Alabama, Georgia, and Florida during 1949. J. Proctor. *Petrol. Tech.*, Dec. 1950, **11** (12), 321–2.—Fourteen dry wildcats, and two dry and four producing development wells were completed in Alabama. The 1949 production was 464,456 brl of oil, from Gilbertown and East Gilbertown.

Sixteen wildcats and six field wells (three producers) were completed in Florida. The Sunniland field produced 441,720 brl of oil.

Six dry holes were drilled in Georgia.

There was geophysical work in each of the three states.

Tables list the 1949 wildcats. G. D. H.

1794. Oil and gas developments in Colombia during 1949. G. F. Dean and G. Barrow. *Petrol. Tech.*, Dec. 1950, **11** (12), 417–21.—In 1949 Colombia produced 30,463,743 brl of oil, a record. This included 819,768 brl of condensate. Output was restricted by strikes and by the capacity of the pipeline in the case of Casabe. Ninety-one development wells were completed; forty-five were at Casabe and forty-one at Tibu. Fourteen wildcats were completed. On test Tetuan No. 1 in the Upper Magdalena Valley gave 744 b.d. Gualanday No. 1 had several shows, but negative results on test. Small shows were noted in Cerrejon No. 1 in the Rancheria basin.

There was surveying activity employing surface geology, gravity meter, magnetometer, seismograph, and core-drilling.

Eight booster stations were under construction along the Andian pipeline. Tables summarize field and wildcat data. G. D. H.

1795. Oil and gas developments in Michigan during 1949. G. E. Eddy and R. P. Grant. *Petrol. Tech.*, Dec. 1950, **11** (12), 329–42.—Of 1004 wells completed in Michigan in 1949, 439 produced oil and ninety-two gas. The footage was 2,242,321 ft. Oil production totalled 16,503,473 brl, and gas 14,576,497 M.c.f. 335 exploratory wells were drilled, thirty-three being successful. Their footage was 720,336 ft. Brine production averaged 172,781 b.d., and 171,240 b.d. was returned to underground reservoirs (mainly Marshall, Traverse, Dundee, and Detroit River formations).

Production statistics are given by fields for oil and gas. Important 1949 wildcats and discovery wells are listed with salient data. Maps and a generalized stratigraphic column are appended. G. D. H.

1796. Oil and gas developments in Mississippi during 1949. H. M. Morse and J. Proctor. *Petrol. Tech.*, Dec. 1950, **11** (12), 323–7.—In 1949 Mississippi produced 37,855,120 brl of oil and condensate from thirty-six fields. The gas production was 108,978,452 M.c.f. About 35,000,000 M.c.f. of gas was injected at Brookhaven and Cranfield.

349 wells were drilled, 112 being wildcats, which found eight oil or gas fields and four salt domes. The oil discoveries were Armstrong Bude, Hartman, Possum Corner, Russum, and Sand Hill. Among the field wells 179 were successful.

Tables give oil- and gas-production statistics by fields, and summarize data on the 1949 wildcats. G. D. H.

1797. Progress report: Romere Pass field. J. A. Kornfeld. *Oil Gas J.*, 1.2.51, **49** (39), 50.—Seventeen producers have been completed in the Romere Pass field on the Mississippi River Delta since it was discovered by 1 North Main Pass Unit in Jan. 1950. Field production is 4300 b.d. from a proven area of approx 760 acres. Fourteen additional wells are planned.

Development of the field, where the ground surface is approx 1 ft above sea-level, involved dredging and the use of submersible drilling barges; a central tank battery, treating and separating equipment, and an oil-barge-loading terminal have been installed.

The field was discovered by gravity and seismic work, and the structure is a deep-seated salt dome with a well-developed shallow graben fault pattern. Two Miocene sand pays at 9000 and 9700 ft are under development; both operate under a water drive. A number of gas-distillate reservoirs have been recognized from sidewall cores.

Drilling and completion practice is described. C. A. F.

1798. Oil and gas developments in Pacific Coast area during 1949. L. E. Porter and L. W. Chasteen. *Petrol. Tech.*, Dec. 1950, **11** (12), 374–98.—California produced 332,942,156 brl of oil in 1949, and 47% came from ten fields. 1723 wells were completed in eighty-nine fields. South Cuyama was opened, and is the best find for ten years. The Placerita field had 228 new wells. Sixteen new fields and pools were discovered.

At Grays' Harbor in Washington a 9344-ft test had showings at several points. A well in Oregon was abandoned at 6480 ft in metamorphics. Several wells were drilled in the Winslow–Holbrook area of Arizona, but the results appear to have been disappointing.

Tables give the 1949 California discoveries, and the main production data by fields. G. D. H.

1799. Pennsylvanian hot spot. Anon. *Oil Gas J.*, 18.1.51, **49** (37), 70.—Leidy gas field, Clinton Country, Pennsylvania, discovered in Jan. 1950, is producing gas from a sand between 5700 and 6400 ft. Gas pressure is about 4200 p.s.i. Tests now being drilled indicate that the currently proved area may be doubled. C. G. W.

1800. Oil and gas developments in the Rocky Mountain region during 1949. R. M. Larsen. *Petrol. Tech.*, Dec. 1950, **11** (12), 343–67.—The Rocky Mountain region produced 81,228,000 brl of oil in 1949. The measured gas production was 92,057,000 M.c.f. 1245 wells aggregating 3,934,489 ft were drilled. Oil was discovered in the Tertiary at Roosevelt, Utah, in the Uinta Basin, and at Gurley, Western Nebraska, in the Denver Basin.

Fifty-five oil wells were completed in Colorado, thirty-nine being in the Mancos shale and nine in the Weber sandstone. Oil was found at Oak Creek and gas at Asbury Creek.

Drilling began on the Big Elk Mountain structure in Idaho. Hitherto forty-one wells have been drilled, most of which were shallow and before 1933. Some found gas in the lacustrine Payette formation.

140 oil wells were completed in Montana. Discoveries were made at Sumatra, Devil's Pocket, and Jack Creek.

One well was under way at Apex in Nevada, where some thirty-four shallow wells had previously been drilled. Three small gas wells were completed near the southern end of the Cedar Creek anticline in North Dakota. Sixty-five tests have been drilled in North Dakota to date, twenty-eight being gas wells at Cedar Creek.

Three wells were abandoned in South Dakota, which has now had eighty-eight tests. Five have given gas–water wells in the Pierre, and there are six low-pressure shut-in gas wells at Ardmore.

Twenty-five oil wells were completed at Ashley Creek in Utah. There were two shallow completions at Boundary Butte.

191 exploratory wells were drilled in Wyoming, and forty were successful. Discoveries included Borie, Glenrock, Hay Creek, Lake Creek, Lightning Creek, Little Sand Draw, Lodgepool, Lonetree Creek, Murphy Dome, North Meadow Creek, Riverton, Sage Spring Creek, Salt Wells, and South Frisby.

Tables giving drilling and production data, and list the fields with salient information. Important wildcats are listed, and a stratigraphic chart is given for Wyoming.

G. D. H.

1801. Oil and gas developments in Hugoton area during 1949. J. F. Dougherty. *Petrol. Tech.*, Dec. 1950, **11** (12), 373.—639 gas wells were completed at Hugoton in 1949. A new gas line to Wisconsin and Michigan was completed. At Richfield gas-condensate was discovered in the basal Pennsylvanian at 5000 ft. Oil was discovered in the Kismet field in the Marmaton at 5100 ft.

Gas production totalled 395,790,000 M.c.f. G. D. H.

1802. Oil and gas developments in Texas Panhandle area during 1949. J. F. Dougherty. *Petrol. Tech.*, Dec. 1950, **11** (12), 368–72.—527 oil wells were completed in the Panhandle field in 1949, and one each at Lips and Palo Duro Creek. Panhandle produced 33,016,852 brl of oil. 131 gas wells were completed at Panhandle, and 833,210,000 M.c.f. of gas was metered. Production of channel black was 376,000,000 lb from 229,000,000 M.c.f. of gas. Furnace black amounted to 90,626,000 lb from 10,300,000 M.c.f. of gas. The Lips field has two Pennsylvania gas reservoirs, and gives oil from the Upper Mississippian limestones.

Palo Duro Creek gives gas from the Lower Pennsylvanian at 5380 ft, and oil at 5900 ft.

The important 1949 wildcats are listed, and salient field data are given. A map is included. G. D. H.

1803. Oil and gas developments in West Virginia during 1949. Anon. *Petrol. Tech.*, Dec. 1950, **11** (12), 398–9.—700 wells were drilled in 1949 (427 gas, seventy-eight oil, twenty-four oil and gas, 144 dry). Gas production was estimated at 181,380,000 M.c.f. and oil at 2,852,500 brl, from 13,800 gas wells and 15,000 oil wells.

A table gives data on drilling and production by counties. G. D. H.

1804. Proved reserves of crude and other liquids continue climb. P. C. Ingalls. *Oil Gas J.*, 25.1.51, **49** (38), 197.—Recoverable crude-oil reserves in the U.S.A. are estimated at 26,121 million brl, of which Texas holds approx 55%, California approx 16%, Louisiana approx 8%, and Oklahoma approx 6%.

Recoverable reserves of natural-gas liquids are estimated at 3,831 million brl, and dry natural gas at 184,945 billion cu. ft. C. A. F.

1805. Oil and gas developments in Brazil during 1949. Anon. *Petrol. Tech.*, Dec. 1950, **2** (12), 405–7.—In 1949 Brazil produced nearly 110,000 brl of oil, but there is a potential of about 1,000,000 brl.

Tables give the products of Brazilian refineries in 1949, the imports and their sources, salient field data, and information about important wildcats. G. D. H.

1806. Oil and gas developments in Peru during 1949. Anon. *Petrol. Tech.*, Dec. 1950, **2** (12), 444–8.—In 1949 Peru produced 14,796,092 brl of oil. 166 development and twenty-six wildcat wells were completed. All the discoveries were on fault blocks adjacent to producing areas.

The proposed new petroleum law has been published, and the principal features of this are summarized.

Tables give 1948 and 1949 production by companies, development and exploratory drilling in the same two years, and provide the salient field data. G. D. H.

1807. Oil and gas developments in Canada during 1949. Geol. Sur. Canada. *Petrol. Tech.*, Dec. 1950, **11** (12), 409–15.—Alberta produced 20,300,000 brl of oil in 1949, and over 800 wells were drilled. Reserves rose to at least 1,000,000,000 brl. 209 wells were drilled at Redwater. Oil was found in the D-3 zone at Golden Spike. 200 miles northwest of Edmonton, Devonian oil was found at Normandville. Other finds were at Barrhead (Mississippian), Bon Accord and Excelsior (Devonian), Joseph Lake, Whitemud, Campbell, and Volmer (Cretaceous).

Ontario produced 265,000 brl of oil, and there was extension of the Becker West field. There was wildcatting in New Brunswick and the Gaspé peninsula.

Important gas finds were made in Alberta at Pincher Creek, Hanna, Castor, and Lac La Biche.

Tables summarize the results of drilling in 1949, give basic data on the Alberta fields, and on the important 1949 wildcats. G. D. H.

1808. Oil and gas developments in the Netherlands during 1949. A. A. G. Schieferdecker. *Petrol. Tech.*, Dec. 1950, **2** (12), 408.—Eighteen producers were completed at Schoonebeck in 1949. Production was 4,316,000 brl.

Sixteen wildcats were drilled, and gas in commercial amounts was found at De Wijk, 25 miles west of Schoonebeck. The Permian gas at Coevorden is to be developed. G. D. H.

1809. Roumanian production. B. F. Linz. *Oil Gas J.*, 15.2.51, **49** (41), 65.—It is reported that Roumanian production has increased by 140% since the industry was nationalized in 1948; actual current production is believed to be about 85,000 b.d. Oil is said to have been found in several areas other than the Prahava Valley, formerly the major producing district. C. A. F.

1810. Austrian production. Anon. *Oil Gas J.*, 8.2.51, **49** (40), 70.—Estimated Austrian production in 1950 was 1,500,000 tons, or approx 28,000 b.d., and current production is believed to be over 32,000 b.d.

Zisterdorf is reported to be producing 22,400 b.d., Matzen, opened in 1949, 20,000 to 30,000 tons, and Aderklaa, where oil was discovered in 1950, 20,000 tons per month. Exploratory work is believed to be in progress in the Bockfliess area.

Seven refineries are operating in the country, five being controlled by the Soviet Mineral Oil Administration, which controls 90% of the Austrian industry. C. A. F.

1811. Oil and gas developments in Egypt during 1949. E. le M. Trafford. *Petrol. Tech.*, Dec. 1950, **2** (12), 422–5.—In 1949 Ras Gharib gave 8,372,038 brl of oil, Hurghada 316,845 brl, Sudr 5,447,652 brl, and Asl 1,670,640 brl. Sudr is virtually fully developed, but Asl is not yet fully defined. Asl has seven producers from a faulted Eocene limestone high covered by Miocene. The limestone porosity is due to weathering as at Sudr and Ras Matarma. The latter originally produced from a mid-Miocene sand, but there is a narrow fault accumulation in the Eocene.

At Wadi Feiram a 120-b.d. well was completed at 6505 ft in the Miocene near to the basement. The possibilities are limited.

Important 1949 wildcats are listed, and a table gives salient data on the fields. G. D. H.

1812. Oil and gas developments in Iraq during 1949. Anon. *Petrol. Tech.*, Dec. 1950, **2** (12), 416.—Four producers and five observation wells were completed at Kirkuk in 1949. Ain Zalah has six producers with a capacity of 30,000 b.d., but no outlet. Two wells were completed on Nahr Umr and Zubair, giving major discoveries on these seismic structures at 8363 and 11,173 ft, respectively. Naft Khaneh has two producers and two observation wells. G. D. H.

1813. Oil and gas developments in Burmah, India, and Pakistan during 1949. Anon. *Petrol. Tech.*, Dec. 1950, **2** (12), 404.—In 1949 there was little progress with rehabilitation in Burma.

At Digboi development was concentrated on the eastern extension, and the output was about 1,950,000 brl. 10 miles south of Digboi exploratory drilling was discontinued because no commercial production was found.

Khaur, Dhulian, Joya Mair, and Balkassar gave 940,000 brl of oil. On the Meyal structure northwest of Dhulian exploration was continued, and the Lakhra well was taken nearly to 12,000 ft without finding oil indications. G. D. H.

1814. Oil and gas developments in the Far East during 1949. Anon. *Petrol. Tech.*, Dec. 1950, **11** (12), 426–32.—In 1949 Indonesia produced 42,221,285 brl of oil. There was surface exploration in North Sumatra, and in Mid-Sumatra, where five producers were completed at Lirik. Twelve wildcats were drilled in South Sumatra, giving an extension to the Radja field and gas discoveries at Lebong and Baung.

British Borneo gave 25,112,322 brl of oil, mainly from Seria. A successful wildcat was drilled at Poeloe Boenjoe in Tarakan. Southeast Borneo gave 3,168,751 brl of oil. Java had considerable surface geological work, and one wildcat, a failure.

Wildcat drilling was in progress in Western New Guinea, but the only completion (dry) was at North Klamono. The 1949 production was 1,743,588 brl. Ten wells were completed in the Klamono reef limestone field.

The Laochunmiao (Yumen) field in Kansu Province, China, produced 2000 b.d.

A well drilled on the Bondoc Peninsula in the Philippines reported encouraging oil flows. A well drilled at Daan Bantayan encountered gas and oil shows. The main points of the new Philippines oil law are given.

In Papua the Oroi well was abandoned, and wells at Upoia and Hohoro reported shallow gas shows.

Geophysical work was in progress in Australia. The Kopper–Amanna test was abandoned in the Jurassic.

Japan produced 1,368,406 brl of oil in 1949, Yabase giving one-third of the total. A new oil horizon was opened at 3800 ft, and others are indicated at 4200 and 5100 ft.

In Assam the Digboi field gave 5500 b.d. Three dry wildcats were completed.

G. D. H.

Transport and Storage

1815. Design and operating considerations of tank-vapor-recovery systems. D. H. Stormont. *Oil Gas J.*, 24.5.51, **50** (3), 98.—Factors in designing a vapour-recovery instillation include volume and specific gravity of gas, dia of piping, pressure drop required, and allowable pressure drop.

Sensitive and reliable vapour-recovery regulators, to work within the range 0·2 inch vacuum to 2 inch pressure (water) are essential.

Small compressors made possible installation of systems which otherwise were not practicable.

This article is the second of a three-part series. G. A. C.

1816. Adequate foundations are first requirements for good tank service. I. E. Boberg. *Refiner*, 1951, **30** (4), 125–7; *Water Tower*, Mar, 1951, **37**, 5; *Oil Gas J.*, 7.6.51, **50** (5), 106.—The desirability and means of providing adequate foundations for large oil-storage tanks are considered. Examples are quoted showing how poor foundations have led to collapse or unsatisfactory tank construction. Sand beds are recommended, with good drainage facilities. Contrary to previous opinion, ring walls for tank support are to be desired. As an illustration, the calculation of a ring wall for a 150 ft dia × 48 ft floating-roof gasoline-storage tank is given. R. W. G.

1817. Storage tank insulation. Dauwalder. *Bull. Ass. franç. Tech. Pétrole*, 1951, (87), 27–31.—Brief description of the Gregson system of protecting lagging by easily removable galvanized sheets. Calculations are shown for determining required lagging thickness in typical cases of fuel-storage tanks. V. B.

1818. Underground storage of a propane–air and natural gas mixture. H. L. Fruechtenicht and J. B. Simpson. *World Oil*, Feb. 1951, **132** (2), 183.—Account is given of the problems and design of the construction of a 10 million cu. ft/day propane–air storage

plant from which the gas was pumped to underground storage reservoirs. The properties of the gas mixtures were investigated with reference to recovery, vaporization, condensation, and hydrate formation. Although operating expenses were relatively high, the scheme was intended primarily as an emergency measure.

A flow diagram and graphical data are given. A. J. H.

1819. Underground storage of propane in a salt water sand. C. B. Johnson. *Petrol. Tech.*, Jan. 1951, **3** (1), 14.—The price per brl of propane has fluctuated seasonally, and there has been a downward trend as output has risen. Propane tank storage of 1000-brl capacity would cost $16·5/brl installed. Propane sold as vapour via gas pipeline might give only the equivalent of 8 c/brl, and the low for liquid sale might be 1 c/gal. At 3½ c/gal and 25% recovery efficiency the payout on a $250,000 investment would be seven years (it being assumed that the initial stored volume underground is 100,000 brl, and losses at 8 c/brl, while the recovery efficiency is that part of the stored material which is recovered).

The Carthage Subsurface Storage Project was planned for a total volume of 150,000 brl, but only one-third of this capacity was utilized from inception to first withdrawal. In the first year of operations over 26·8% of the stored propane was marketed.

G. D. H.

1820. The story of the Texas-Illinois Natural Gasoline Pipeline Co. J. Oates *et al.* *Oil Gas J.*, 3.5.51, **49** (52), 81.—The new Big-Inch Gulf Coast to Chicago pipeline system is reviewed in eight sections; dealing respectively with its overall purpose and service objective, design capacity, its builders, pipe-laying methods, river crossings, communications, selected route, and compressor-station design. G. A. C.

1821. Economics of increasing capacity of pipeline systems. N. B. Mavris. *Oil Gas J.*, 17.5.51, **50** (2), 119.—Original design of pipeline should provide for eventual capacity in most economical manner. Curves have been plotted for a comparison of looping *v.* booster stations from a standpoint of savings in operating costs against required additional investment, on single-line systems.

Ten curves show that if increase in capacity is to be above 50%, loops are economical, but if below 50% and stations are more than 50 miles apart, boosters are more advantageous; but loops should be used for increase below 50% with stations less than 50 miles apart, but actual saving is small in the latter case. G. A. C.

1822. Lightweight pipelines: aluminium has come of age. W. B. Moore. *Oil Forum*, May 1951, **5** (5), 162.—This paper is the continuation of one in the Feb. issue of *Oil Forum* (Abs. 1221). During the extension of the natural-gas-distribution system of Alabama & Tennessee Natural Gas Co. to the Listerhill Plant of Reynolds Metals Co., a 1·8-mile section of 8⅝ inch-O.D. aluminium pipe was incorporated with the steel line. Itemized installation costs showed savings of $0·166/ft of pipe laid over the costs for steel pipe. In other words, a saving of 21·4% for aluminium. If coatings had not been applied, the installation cost would have been 32·6% lower than that of the steel line.

For material-cost comparison, the steel price is based on $135/ton, making the aluminium pipe cost 55·4% more than the steel.

Based on the extensive test programmes which have been undertaken, the advantages of aluminium pipelines are: (1) Lower handling costs. (2) Elimination of wrappings in many types of soil. (3) Elimination of pipe cleaning, where wrapping is required. (4) Better corrosion resistance and longer life. (5) Elimination of painting in open installations. (6) Reduced welding costs. (7) Elimination of heavy suspension in overhead systems. (8) Decreased pumping costs, due to lower coefficient of friction. (9) Higher scrap-recovery value. A. S.

1823. Methods of absorbing expansions of long-range transmission piping. F. E. Wolosewick. *Refiner*, 1951, **30** (2), 69–74.—The merits and uses of expansion loops and expansion joints are considered, together with the problems associated with branch piping, buried lines, and maintenance. Generally, expansion joints are not necessary to absorb expansions of considerable magnitudes—use being made of the flexible properties of weld ells and bends—although where pressure drop is important

expansion joints may be used with advantage. Reference is also made to the conditions of pre-straining, so as to put process equipment under no strain during operation. R. W. G.

1824. Pipe fluid flow friction loss determinations. E. M. de Forest. *Refiner*, 1951, **30** (3), 97–106.—To avoid the necessity of performing time-consuming calculations for routine flow problems, a method entailing the use of much graphical data is presented. The method briefly consists of applying a correction factor obtained from a knowledge of kinematic visc to pressure loss in pipes for water. A large number of pipe sizes are considered for clean steel. For other pipes a correction must be made. Values obtained by this rapid method are in very close agreement with calculated values. R. W. G.

Refinery Operations

Refineries and Auxiliary Refinery Plant

1825. New refinery in operation at Shell Haven, Essex. V. S. Swaminathan. *Refiner*, 1951, **30** (2), 78–80.—A review of the new Shell plants in England which presents many illustrative figures on the U.K. petroleum-refining capacity. The capacity is being increased from 18·5 to 145 million brl over the years 1947–53. In 1952 Shell's U.K. capacity will be 48 million annually—proving the largest single output in the country. The capital cost of the works planned at Shell Haven will be some $34 million. Reference is made to the various processes employed by Shell. R. W. G.

1826. Shell's Provident City gasoline plant in full operation. Anon. *Refiner*, 1951, **30** (4), 84.—Following the completion of a 110-mile 16-inch pipeline to carry gas to Baytown, Texas, Shell's gasoline plant at Provident City, Texas, is treating 60,000,000 cu. ft/day of raw gas. Residue gas is dehydrated for pipeline sales, while approx 2500 b.d. of liquid products are yielded. R. W. G.

1827. New British American refinery is complete combination plant. Anon. *Refiner*, **30** (4), 128.—An advanced example of combination processing is being erected for the British American Oil Co. Ltd., at Edmonton, Alberta. The refinery is a complete combination plant operating off one central control room. Processing includes crude distillation, delayed coking, thermal and fluid cat cracking, gasoline stabilizing, light-ends fractionating, and light-oils treating. The first of its kind, the refinery will process 2,000,000 brl/year. R. W. G.

1828. Llandarcy refinery expansion work now in second phase. V. S. Swaminathan. *Refiner*, 1951, **30** (5), 96–7.—A review of the present expansions of the Llandarcy refinery, South Wales. When work is completed the considerably enlarged refinery will produce a comprehensive range of products including propane and butane, premium gasoline, regular gasoline, kerosine, T.V.O., various grades of gas oil, a complete range of solvent-refined lubricating oils, including bright stocks, various grades of wax, fuel oil, and bitumen. New units include plant for SO_2 extraction of kerosine, solutizer for benzine, desulphurizer for T.V.O. and gasoline, and several acid washers and re-run units. R. W. G.

1829. Process systems applicable to military aviation fuel manufacture. D. Read. *Refiner*, 1951, **30** (3), 130–6 (*Western Petroleum Refiners Association mtng, Lake Charles, La.*).—A review of the possible ways of producing high-grade gasoline for reciprocating aviation fuels during a military emergency. Processes discussed are cat cracking and reforming, cat polymerization, HF alkylation, platforming, and aviation-gasoline blending. Conclusions reached are that cracking of heavy gas oils rather than light stocks is preferable in view of the probable demand for jet fuel, highly olefinic cat-cracked products should be platformed, platforming should be used to relieve aromatic shortage, and increased use of platforming should reduce the necessary quality of alkylate, thus allowing utilization of propylene, amylenes, cat butylene polymers, and butylene as feedstocks. R. W. G.

1830. Panoma plant achieves low maintenance-operation costs. Pt I. I. E. Nutter and C. D. Van Vliet. *Refiner*, 1951, **30** (4), 71–6.—A general description of the Panoma Corporation's gasoline plant near Hooker, Texas Co., Oklahoma, including such features as choice of plant site. It is claimed that as a result of the original design considering operating and maintenance costs, performance has been excellent. A line diagram of this 100,000,000 s. cu. ft/day raw-gas plant shows the use of rich oil rectification without the use of compressors to give a high propane recovery. R. W. G.

1831. Panoma plant achieves low maintenance-operation costs. Pt II. I. E. Nutter and C. D. Van Vliet. *Refiner*, 1951, **30** (5), 98–102. (See preceding Abs.).—Pt I gave a general description of the process details of the Panoma Corporation's gasoline plant near Hooker, Texas Co., Oklahoma. Pt II discusses the auxiliary equipment for this unit, which is claimed to have been successfully designed to give low maintenance and operation costs. R. W. G.

1832. Taylor–Mayfair cycling plant achieves efficiency with simplicity. L. Gross and G. T. Wells. *Refiner*, 1951, **30** (2), 115–19.—A description of the latest cycling plant of Taylor–Mayfair, N. R. McAllen, Texas. Processing capacity is 75 million cu. ft/day, but is to be doubled. Fractionation, rather than stripping, provides the removal of methane from rich oil, and for the retention of desired hydrocarbons in the rich oil. R. W. G.

1833. Maintenance facilities in gasoline cycling plants. H. Givens. *Refiner*, 1951, **30** (4), 77–80.—The maintenance crew requirements are considered and numbers of different workers required are suggested—*i.e.*, number of electricians, pump mechanics, etc., required. A type of form useful for keeping records is indicated. R. W. G.

1834. Portable low temperature processing equipment. G. F. Russell. *Refiner*, 1951, **30** (4), 81–3.—An economically feasible portable unit employing the refrigerating potential of expanding gas is described, which is suitable for handling small quantities of gas ahead of pipeline sales. One of the units described has a capacity of 3,500,000 s. cu. ft/day of raw gas. The advantages obtained are increased hydrocarbon condensate or gasoline recovery and greatly reduced water content of effluent gas. R. W. G.

1835. Refrigeration in the refining industry. Pt I. T. G. Hicks. *Refiner*, 1951, **30** (3), 88–90.—The first article of the series discusses generally the compression (reciprocating and centrifugal), absorption and steam-jet vacuum systems. All systems are not completely interchangeable, for choice depends on the operating conditions. R. W. G.

1836. Performance of cooling equipment. H. A. Dresser. *Refiner*, 1951, **30** (5), 110–13.—A discussion of cooling systems, laying emphasis on regular inspection to assure good performance. Particular reference is made to the nature of water used—control should be effectively made of its pH and other properties liable to affect coolers. Several suggestions are offered on the aspect of regular inspection. R. W. G.

1837. Use of aluminium in petroleum refinery equipment. E. E. Kerns and W. E. Baker. *Refiner*, 1951, **30** (5), 123–9.—A series of tests and operation data on aluminium in use on petroleum plant is presented, indicating the possibilities of using aluminium fabrication. Aluminium is resistant to corrosion by sulphur, organic acids, ammonia, and furfural, but is anodic to most other common materials of construction or their salts. Hence it is necessary to design installations so that certain ions, such as lead, copper, mercury, and chlorides, are not present. It is shown that aluminium may be advantageously used in many instances—*e.g.*, for tank roofs and in furfural refining. R. W. G.

1838. Pressure drop through porous media IV—New data and revised correlation. L. E. Brownell, H. S. Dombrowski, and C. A. Dickey. *Chem. Engng. Prog.*, Aug. 1950, **46** (8), 415–22.—A continuation of investigations reported in *Chem. Engng Prog.*,

1947, **43**, 537 [Abs. Nos. 1764–6, 1948]. New data on flow through porous media are presented for packed beds covering a range from completely laminar to highly turbulent flow, and a porosity range from 40 to 94%. Observed pressure drops are compared with those predicted by various correlations, and the method of correlation presented in Pt I of this series is revised. J. G. H.

1839. Installation, operation, maintenance of mechanical drive steam turbines. W. B. Yard. *Refiner*, 1951, **30** (4), 104–8.—Steam turbines are in relatively large use, and many operators have definite opinions as to proper procedure to be followed. Manufacturers' opinions are presented. Emphasis is laid on correct installation such that the unit is in line under working conditions and free from stress. Notes on operation and maintenance are given which should prove useful to operators. R. W. G.

1840. Preventive maintenance for engines and compressors. R. S. Ridgeway. *Refiner*, 1951, **30** (4), 64–70.—A discussion of the advantages of regular maintenance of gas engines and compressors. By establishing periods of safe operating time it is possible to reduce maintenance costs, have smaller crews, and improve performance. Discussion also includes notes on looking for engine wear during operation by using indicator diagrams, etc. Notes on scheduling maintenance and keeping records are given. R. W. G.

1841. Process steam generated by using waste heat. J. C. Albright. *Refiner*, 1951, **30** (3), 119–21.—The method of utilizing waste heat to generate process steam on a gasoline plant is described. A waste-heat boiler effectively uses the heat of the lean absorber oil to supply steam for stripping the rich oil. Production of this process steam is automatic and inexpensive. R. W. G.

1842. New development in radiant heating for the petroleum industry. C. C. Tate. *Refiner*, 1951, **30** (5), 103–6 (*Western Petroleum Refiners Association, San Antonio*, 1951).—Use of a flameless ceramic cup burner as a radiant-heat source for refinery tube stills is described. This type of burner enables good control to be effected over the time–temp curves. Operation of a furnace using these burners has been closely followed, and typical conditions give: (1) average radiant-heat transfer rate of 17,000 B.Th.U/sq. ft/hr; (2) average convection heat transfer rate of 4,920 B.Th.U/sq. ft/hr; (3) heater efficiency of 81·5%; and (4) excess air in stack 9·2%. Favourable results are obtained with this heater in gas oil cracking, for both catalytic cycle oil and virgin gas oil, giving higher octane ratings than normal, and in visbreaking of high vacuum petroleum residuum. The still is not susceptible to excessive coking. R. W. G.

1843. Improvised sulphur burner saves half billion gallons per year of tower blowdown. V. F. Estcourt. *Oil Gas J.*, 14.6.5, **50** (6), 105.—A sulphur burner is described in which 100 lb sulphur is burnt, the SO_2 liberated being drawn into the cooling tower and absorbed by water falling over the baffles. This considerably reduces amount of scale deposits on condenser tubes, giving a commercially clean condenser. Supplementary chlorination prevents algæ growth. G. A. C.

1844. Graphical methods aid in stack foundation design. C. H. Fork. *Refiner*, 1951, **30** (3), 81–5.—A procedure is developed for the direct calculation of foundation size for stacks and elevated tanks, allowing for lateral pressure due to wind or an earthquake and soil-bearing values. The method, which is developed mathematically from simple mechanics, is assisted by means of graphs. R. W. G.

1845. Maintenance and operation—How to do it. Anon. *Refiner*, 1951, **30** (2), 87–9.—A set of practical notes on how several small but important maintenance jobs may be accomplished. For example, a simple under-water viewer is described which may be used for locating submerged tools or in tightening leaking flange bolts. R. W. G.

1846. Maintenance and operation—How to do it. Anon. *Refiner*, 1951, **30** (3), 115–17.—A further set of practical notes on how several small but important maintenance jobs may be accomplished. For example, development of a new method of removing exchanger bundles results in lower initial investment for supporting structures in processing plants. R. W. G.

1847. The application of centrifugal pumps. A. E. Harnsberger. *Refiner*, 1951, **30** (1), 119–28.—The article is meant to present details of performance of centrifugal pumps to enable sufficient to be understood in choosing or suggesting actual designs. Data on capacity, head, and efficiency are given graphically. Emphasis is placed on the conditions of suction to the pump. R. W. G.

1848. Centrifugal pumps in chemical industry. J. Gahus. *Chim. et Ind.*, 1951, **65**, 657–64.—A discussion of mechanical seals, with drawings of five patterns (four of them French). Lubrication is particularly considered, especially the pressure-feeding of the lubricant; the non-admixture of lubricant and liquid pumped is demonstrated. V. B.

1849. Chemical treating plant for refinery waste water. H. Schindler. *Oil Gas J.*, 31.5.51, **50** (4), 92.—The treating plant at the Daugherty Refinery of Sonneborn Sons is described.

Cooling water from sludge-conversion unit, waste water from manufacture of white oils and recovery of petroleum sulphonates make up bulk of water to be treated.

The waste water is adjusted to a pH of 5, hydrated lime at pH of 7·0 to 8·4 added to precipitate sulphonic and mineral acids, the precipitate continuously settled, and treated water discharged.

Tables give properties and volume of waste water, and operating data of the treating plant for six representative months. G. A. C.

Distillation

1850. Industrial fractionating tower packing. R. C. Scofield. *Chem. Engng Prog.*, Aug. 1950, **46** (8), 405–14.—Performance characteristics are reported for a new type of industrial fractionating-tower packing, known as Series No. 7, having both higher capacity and "H.E.T.P." when compared with conventional bubble-cap towers. Packing is fabricated to form corrugated trays of multilayer expanded metal lath, the trays being so assembled as to bring the ridges and valleys of adjacent trays into contact. Experimental performance characteristics under a variety of conditions are detailed, together with commercial applications for towers up to 4 ft in dia. Substitution of the new packing for thirty-six bubble-cap trays on 1·5 ft spacing results in a performance equivalent to more than 110 bubble-cap trays while operating at twice max original loadings. Savings reflecting up to a sixfold reduction in tower size are claimed, with other associated cost items. J. G. H.

1851. New flash tower resists high sulphur corrosion. L. W. Williams. *Refiner*, 1951, **30** (1), 144.—The refining of deep-well sour crudes leads to straight crudes containing upwards of 2½% sulphur. To the corrosive attack of this element must be added that caused by the formation of hydrochloric acid and organic chloride. At Socony-Vacuum Oil Company's plant, Paulsboro, N.J., the plant is protected by stainless-clad steel, type 405. R. W. G.

1852. Low pressure distillation. J. Jaeckel. *Erdöl u. Kohle*, 1951, **4**, 175–6.—Molecular, free-path (v.p. $\geqslant$ residual gas pressure) and steam dist processes are compared from the installation and economical points of view. It is deduced that free-path dist is more efficient than molecular dist, both processes requiring preliminary de-gassing of feed to column. Steam dist is better suited for higher pressures than free-path dist, the latter being preferable at pressure < 2 mm. Description of the Herlet apparatus for determination of v.p. and mol. wt. is given. Application of high vacua to large-scale processes is discussed, with special reference to the different types of evacuating equipment and the problem of leakage and its prevention. P. V.

1853. The evaporation of droplets in still air. G. O. Longstroth, C. H. H. Diehl, and E. J. Winhold. *Canad. J. Res.*, Nov. 1950, **28A**, 580–96.—The evaporation of droplets of 1 to 2 mm dia into still air has been studied with nine liquids forming a series of graded volatilities from toluene to aniline and including water. The experimental results agreed with calculations based on diffusion and heat-transfer theory. The surface temp of the more volatile droplets was found to be significantly lower than the bulk

temp in contrast to Johnson's results with water. Convection effects were found to be present with 43 mm dia evaporation vessels—confirmation of Gilliland's suggestion, but were absent in the smaller vessels and in the main investigation. N. T.

1854. Applications of diffusion theory to evaporation from droplets and flat surface. G. Luchak and G. A. Longstroth. *Canad. J. Res.*, Nov. 1950, **28A**, 574–80.—It has been customary in applications of diffusion theory to evaporation problems to consider the liquid surface as fixed in solving for the vapour distribution in the adjacent space. A more rigorous treatment, involving solution of the diffusion equation with moving boundaries has been applied to the problem of evaporation from droplets and flat surfaces. The results indicate that the equations obtained by the method of quasi-stationary states are good to a high degree of approximation in ordinary circumstances. N. T.

1855. A system of process calculations for light hydrocarbons. Pt V. Fractionation. A. J. L. Hutchinson. *Refiner*, 1951, **30** (2), 107–12. (See Abs. Nos. 658, 1033, 1421, 1861.)—The series is concerned with a method of using printed forms to expedite the work of process-plant calculations. Pt I treated the basic principles, Pt II vapour–liquid flash computations, Pt III absorption, and Pt IV stripping. Pt V is concerned with fractionation of light hydrocarbons (C_1 to C_6), and an illustrative example (using printed forms) is presented. R. W. G.

1856. A system of process calculations for light hydrocarbons. Pt VI. Fractionation (continued). A. J. L. Hutchinson. *Refiner*, 1951, **30** (3), 107–14.—In this article the relation between reflux ratio and No. of theoretical plates is considered, together with heat requirements. An illustrative example is presented. R. W. G.

1857. A system of process calculations for light hydrocarbons. Pt VII. A. J. L. Hutchinson. *Refiner*, 1951, **30** (4), 119–22.—Column sizing is considered. A graph is presented giving surface tension of normal paraffin hydrocarbons plotted against molecular weight for different temperatures. Typical forms are indicated for use in selecting absorbers, fractionators and stills, and for specifying pumps and heat exchangers. R. W. G.

1858. Control of pH in stills with boiler blow-down water. Anon. *Refiner*, 1951, **30** (2), 90–1.—A brief description of the method employed to reduce corrosion by increasing the pH in the water legs on the columns of the gasoline plant of Wilmington Gasoline Co., Terminal Island, Calif. Boiler blow-down water, after filtering and settling, is injected to increase the pH in column water legs from 5·8–6·2 to 6·5–7·5. R. W. G.

Absorption and Adsorption

1859. Dynamic adsorption of ammonia, of butane, of cyanogen chloride and of water vapour by charcoals. S. G. Davis, M. R. Foran, J. D. B. Ogilvie, J. A. Pearce, and C. A. Winkler. *Canad. J. Tech.*, 1951, **29**, 190.—Apparatus designed for continuously determining the wt of gas or vapour removed from an air stream by a porous adsorbent. Evaluated dynamic adsorption of the above four compounds on a steam-activated coco-nut shell charcoal and also dynamic adsorption of cyanogen chloride and of water on steam-activated copper-impregnated coal and wood charcoal; and on steam-activated, copper- and chromium-impregnated nutshell charcoal. Generally results obeyed present-day theories as on adsorption isotherms, etc. The four compounds investigated were all absorbed differently on the coco-nut-shell charcoal butane as a monomolecular layer, ammonia as a gaseous film, cyanogen chloride intermediate between these two, and water was adsorbed by capillary condensation. A. D.

1860. Glycol contamination in gas-dehydration plant operation. J. R. Mazelli. *Oil Gas J.*, 14.6.51, **5** (6), 98.—Diethylene glycol plants suffer fouling of glycol from entrance of extraneous matter with gas stream to contactor, and by corrosion of plant facilities, and from degradation of glycol itself. Data are presented from tests on glycol from the Southern California Gas Co.'s Station 50, tracing the cause of the degradation of the glycol and showing that reclamation can be effected by distillation. G. A. C.

1861. A system of process calculations for light-hydrocarbons. Pt IV—Stripping. A. J. L. Hutchinson. *Refiner*, 1951, **30** (1), 145–8. (See Abs. Nos. 611, 1033, and 1421.)—The series is concerned with a method of using printed forms to expedite the work of process-plant calculations. Pt I treated the basic principles of light-hydrocarbon process-plant calculations, Pt II dealt with vapour–liquid flash computations, Pt III dealt with absorption, while Pt IV deals with stripping. R. W. G.

Solvent Extraction and Dewaxing

1862. Selective solvent refining of crude oil residues using furfural and cresol. O. Geschwind. *Nafta*, Jan.–Feb., 1950, **6**, 20–2.—Residue of SG 927 at 15° C, V.I. 62 pour point −21° C, Conradson carbon 2·6% was refined using anhydrous furfural and using cresol/water mixtures in 90/10 and 95/5 ratio all in 3 × 100% lots.

The anhydrous furfural and 90% cresol gave similar quantities and qualities of raffinate. Yields were approx 56 to 57%, whereas 95% cresol gave a yield of 37% but of much higher-quality lubricant. Results are tabulated. M. S.

Cracking

1863. Atlantic refining announces new catalytic reforming process. Anon. *Refiner*, 1951, **30** (3), 86–7.—Data obtained on the Atlantic Refining Co.'s recent pilot plant for reforming are given. It is claimed that the new pelleted catalyst is particularly suited to reforming low-octane naphthas and producing highly concentrated aromatic stocks. Figures showing degrees of conversion in reforming and aromatic production are given. Features of the process include non-necessity of catalyst regeneration for feeds lighter than kerosine, non-sensitivity to normal water and sulphur concentrations, and production of stock free from sulphur and olefins. Hydrogen recycling is employed. No details of the catalysts are given. R. W. G.

1864. Two new air-lift type TCC units put into operation. Anon. *Refiner*, 1951, **30** (3), 95–6.—A plant at the Beaumont refinery of Magnolia Petroleum Co. and one at the Cleveland, Ohio, refinery of the Standard Oil Co. (Ohio) went on stream in January 1951. Both plants have graphic control panels. R. W. G.

1865. Catalytic cracker, other construction progressing at Wilton. Anon. *Refiner*, 1951, **30** (1), 136–7.—A brief review, including illustrative photographs, of the new Imperial Chemical Industries Ltd. plant at Wilton, North Yorkshire. The work is based on two major activities (1) the cracking of oil for the manufacture of organic chemicals, (2) the electrolysis of brine for the production of chlorine and caustic soda. Pipelines nearly 10 miles in length connect Wilton Works to the I.C.I. plant at Billingham. The cost of Wilton Works will be about £22 million.

Wilton will have the first plant to produce the new synthetic fibre "Terylene." R. W. G.

1866. Economics of petroleum coking for catalytic cracking feed preparation. J. W. Jewell, Jr., and J. P. Connor. *Refiner*, 1951, **30** (2), 75–7 (*Oklahoma State Mtng, Amer. Inst. Chem. Engrs, Tulsa*, Oct. 1950).—The economics of delayed coking are discussed by making comparisons, based on complete refinery operation, with three other feed preparations (vacuum distillation, propane decarbonizing, and visbreaking). Delayed coking provides a method of removing the heavy asphaltic materials in a manner enabling removal without interruption of the continuity of the process. The need for coking has been determined by the oversupply of residual fuel oil. Delayed coking plants are reported to have operated at a stream efficiency in excess of 90% for a period of over one year. Present pilot-plant work on the simultaneous production and removal of coke in a system analogous to fluid cat cracking should remove much of the operating prejudice against coking. The summaries presented indicate that, in general, the delayed-coking process has a superior earning power in a soft fuel-oil market or a good coke market. R. W. G.

1867. Visbreaking high-vacuum residuum. J. G. Allen, D. M. Little, and P. M. Wadill. *Oil Gas J.*, 14.6.51, **50** (6), 78.—Phillips Petroleum Co. are developing a high-vacuum distillation process operating at low absolute pressures of 0·1 to 0·8 mm for

the recovery of greater volumes of high-boiling hydrocarbons of low carbon residue. The residuum from the high-vacuum process is then visbroken in a new design radiant heater.

Initial visbreaking operations were subject to frequent shutdowns due to coking of oil and transfer line, but a reduction of coil-outlet temperature on the atmospheric preheater from 835° to 775° F successfully overcame the difficulty. Pilot- and commercial-plant tests of the new process show greater recovery of high-boiling gas oils suitable for catalytic cracking feed stocks. A flow sheet is given; plant conditions and product characteristics are tabulated. G. A. C.

Hydrogenation

1868. Catalytic benzole refining under pressure. W. Urban. *Erdöl u. Kohle*, 1951, **5**, 279–82.—Main disadvantages of old H_2SO_4 benzole-refining process are small yields (approx 85%), difficulty of removing S and S compounds, undesirable side reactions, acid sludge disposal. To avoid these cat process has been developed based on hydrogenation of unsaturated constituents of crude benzole and complete S removal. Process was first patented in 1925, but successful large-scale operation was not achieved till 1950. Original plant capacity of 5000 to 7000 tons/month is to be doubled by May 1951. Comprehensive comparison is made between the two processes. New process gives yields of 98% (wt) of S-free raffinates (65% benzole, 15% toluole, 5% xylole, 9% alkylated benzenes, 4% light and wash oils). Process is independent of feed quality, while its main product is directly marketable as a motor fuel. By simple fractionation a variety of technical products are obtained of a purity unattainable with H_2SO_4 refining. Illustrated description of process and relevant operating details are given. Economics of cat pressure hydrogenation are briefly compared with H_2SO_4 refining, and it is concluded that the vastly larger outputs and decreased production costs of the former must lead ultimately to a complete displacement of the latter. P. V.

1869. Oil hydrogenation in Germany. M. Pier. *Erdöl. u. Kohle*, 1951, **5**, 277–9.—Western Germany has four coal-hydrogenation plants with former total oil output of approx 1 million tons/year. Post-war reconstruction plans were made for utilizing the hydrogenation plants for processing pet, and two plants are now handling crude. Pet is much richer in H than coal, hence much higher process efficiencies are achieved. Former coal and tar hydrogenation practice is closely followed, process consisting of straight crude dist followed by two-stage cat pressure hydrogenation of residuum (b.p. $> 350°$ C). Overall liq fuel yields are approx 95% wt. Unmarketable condensation products are avoided, nature of crude has little effect on process or products, and suitability of cheaper crudes is of great importance to crude-importing countries. Possible combinations of hydrogenation with thermal and cat cracking processes are discussed. Recent developments and some known techniques for improving yields are mentioned. P. V.

Polymerization

1870. Study of the copolymerization of butadiene and styrene in emulsion at subfreezing temperatures. R. J. Orr and H. L. Williams. *Canad. J. Tech.*, 1951, **29**, 29.—The characteristics of this copolymerization were studied at $-18°$ C and $-10°$ C. Polymerization charge recipes have been developed which should permit commercial production at these temperatures at rates comparable to those used at 50° C. The effect of different variables on the rate was determined. Methanol was a satisfactory anti-freeze agent, and either potassium hydrogenated fatty acid soap or hydrogenated resin soap was a suitable emulsifying agent. The latter has to be used in a special "methanol first" technique which is described, and in both cases the pH had to be carefully adjusted. Mixtures of the two soaps were very good. Electrolytes as visc-reducing agents retarded the reaction at low conversion, but caused a higher conversion overall. Para-substituted amine hydroperoxides were compared, with the result that improved performance was obtained as the para substituent became more electro positive. Activators for the redox system were found to be best when composed of a carefully chosen combination of ferrous sulphate, arsenious oxide, potassium pyrophosphate, and tetrasodium ethylene diamine acetate. A. D.

1871. The compositional heterogeneity of butadiene–styrene copolymers synthesized at −18° C in emulsion. R. J. Orr and H. L. Williams. *Canad. J. Chem.*, 1951, **29**, 270.—From analyses for bound styrene, reactivity ratios have been calculated and are compared with figures for reaction at 45° C. Polymers were found to be remarkable homogeneous at low conversions. Number average and visc average mol. wt. were calculated from mercaptan disappearance and vistex intrinsic visc data respectively. The mol.-wt. heterogeneity increased with increasing conversion and initial mercaptan content. The increment number average mol. wt. were found to diminish with conversions, where the increment visc average increased at higher conversions as conversion increased. A. D.

Alkylation

1872. Improvements in sulfuric acid alkylation process. Anon. *Refiner*, 1951, **30** (2), 93–4 (*Condensation of the Kelloggram*, 1950 *Series, Issue No. 5.—The M.W. Kellogg Company*).—The international crisis and increased quality standards for aviation and motor gasoline have led to further interest in sulphuric acid alkylation. A design of reactor is described which maintains an approximately nine-to-one *iso*butane-to-olefin ratio (a desirable condition) within the agitated reaction sections, with a feed of less than four-to-one ratio. R. W. G.

Chemical and Physical Refining

1873. The selection of treating processes for gasoline plants. G. M. Brooner. *Refiner*, 1951, **30** (4), 94–9.—A useful discussion of the removal or conversion of sulphur compounds in gasoline plant products. The conclusions reached are that the most practical and economical treating systems will include: (1) removal of H_2S from gas prior to absorption; (2) a caustic or amine solution wash on the propane LPG production, and (3) copper sweetening of the debutanizer feed or, if high mercaptan content, a combination of regenerative caustic treating and copper sweetening.

Emphasis is laid on the use of amine solutions for removal of H_2S from liquid products so as to lessen the disadvantages of caustic washing, which include high cost and difficult disposal problems. Where mercaptans and other sulphur compounds are present in large amounts a catalytic desulphurization might be justified. R. W. G.

1874. Extraction of mercaptans by means of adsorption. W. E. Voreck, Jr., W. C. Carroll, and P. Zurcher. *Refiner*, 1951, **30** (3), 126–9.—Laboratory and pilot-plant removal of mercaptans from sour distillates by means of a percolation process is described. Adsorbents impregnated with sodium hydroxide solution are used, being regenerated with steam or with steam and air. Only electropositive or neutral adsorbents, such as certain carbons, devolatilized petroleum coke, phenolformaldehyde resins, or basic exchange zeolites, are suitable with sodium hydroxide. For straight-run gasolines, rates of up to 6000 b./ton of adsorbent were used before the treated distillate proved sour to the doctor test. The process depends on the sodium mercaptide being adsorbed, which disturbs the equilibrium between the aqueous and hydrocarbon phases in favour of sulphur removal. R. W. G.

1875. Sweetening and desulfurization of light petroleum products. Pt III. V. A. Kalichevsky. *Refiner*, 1951, **30** (1), 129–35. (See Abs. Nos. 1042 and 1476.)—Plumbite methods of sweetening are discussed in some detail, including both continuous and batch processes. Where elemental sulphur is present in excess, it may be removed by means of sodium plumbite solution and light mercaptans—but the widely used method employs sodium sulphide, which under proper conditions is capable of absorbing elemental sulphur by forming sodium polysulphides. In some instances the undesirable effects of free or loosely combined sulphur may be counteracted by the use of suitable addition agents, such as tetrahydronaphthalene, pinene, or *cyclo*hexene. Petroleum distillates employed in point manufacture and containing corrosive sulphur may be utilized without discolouring the paint upon addition of very small quantities.

(0·001 to 0·05%) of thiophenol, thiocresol, and similar aromatic mercaptans. It is pointed out that the presence of free sulphur in products is normally due to over-addition of elemental sulphur in doctor treatment or the oxidation of H_2S. Both these causes may be avoided.

140 references to the literature are given. R. W. G.

1876. Sweetening and desulfurization of light petroleum products. Pt IV. Hypochlorite treatment. V. A. Kalichevsky. *Refiner*, 1951, **30** (2), 95–6.—Hypochlorite sweetening, employing sodium hypochlorite or calcium hypochlorite is discussed. Hypochlorite treating, because of the possibility of oxidation and/or chlorination of unsaturates, is only applicable to stable petroleum distillates. The chief use is in treating natural gasoline—although it is not of universal use. Fifty-five references given. R. W. G.

1877. Sweetening and desulfurization of light petroleum products. Pt V. Sweetening with copper chloride. V. A. Kalichevsky. *Refiner*, 1951, **30** (3), 122–5.—The refining properties of copper are discussed generally, and with particular reference to copper chloride. Copper easily reacts with sulphur, and may be used in commercial sweetening. Owing to the cost of copper salts, the reagent must be recovered in order to make the process economical. This is usually done by blowing with air. Sweetening with copper chloride depends on the formation of copper mercaptides with simultaneous liberation of hydrochloric acid. The mercaptides react with the acid and oxygen to give copper chloride, disulphides, and water. Copper chloride sweetened products may retain traces of copper which contribute to their instability. Various chemicals may be added to overcome this—such as 0·0001 to 0·002% of *N*:*N*-disalicylidene-1 : 2-diaminopropane. A list of seventy-nine references is given. R. W. G.

1878. Sweetening and desulfurization of light petroleum products. Pt VI. Alkaline reagents. V. A. Kalichevsky. *Refiner*, 1951, **30** (4), 111–18.—The desulphurization abilities of alkaline reagents, including oxides and hydroxides of alkali and alkali-earth metals and their salts of weak acids, particularly carbonates, are discussed. Those mentioned specifically include sodium and potassium hydroxides, ammonia, and alkali-earth compounds. Various organic solvents (solutizers) may be added to caustic soda solutions to increase the ease of removal of mercaptans from petroleum stocks. A list of over fifty solutizers is given, including tannic acid. Several oxidation catalysts for use in the regeneration of spent solutizer solutions are listed, the latest of these being sulinic acids of indigo. 220 references. R. W. G.

1879. Sweetening and desulfurization of light petroleum products. Pt VII. Vapour phase desulfurization. V. A. Kalichevsky. *Refiner*, 1951, **30** (5), 117–22.—This article explains the mechanism and chemistry of the reactions involved in splitting of sulphur compounds at elevated temperatures. A comprehensive list of desulphurizing agents is given. Ninety-two literature references. R. W. G.

1880. Sodium plumbite treating methods and equipment. L. S. Daniels. *Refiner*, 1951, **30** (1), 138–42.—Briefly reports operating procedures and process equipment used by some plants to lower the mercaptan content of light distillate products made from sulphur-bearing crude oils. It might be said that the procedures described here are "typical" of present practice, but variations of the basic methods are usual rather than exceptional, and the better comment is that many doctor-treating schemes follow the general principles described here, if not all the details. (Author's abstract.) R. W. G.

1881. Laboratory procedures used for studying the chemical activation (acid or alkaline) of clays. L. Deibner. *Chim. Anal.*, 1951, **33**, 18–23, 51–8, 95–100, 128–42.—Theory of acid activation is reviewed, and susceptibility of various clays to activation discussed. Methods for the determination of $CaCO_3$, moisture, sand, and other impurities are outlined. Activation by H_2SO_4 or HCl may be used, with former, HCl pre-treatment is desirable to remove Ca. Suitable apparatus and conditions for acid activation are described. Acid concn is less important than total quantity (*i.e.*, on anhydrous

basis) of acid used. Generally acid required is equivalent to approx 25% of anhydrous acid, with 3-hr contact at 80° to 105° C. Acid expenditure is reduced by pre-treatment of clay by electro-dialysis. Washing procedures and control tests are given. Drying is at $\geq 100°$ C to a residual water of 5 to 10% (7 to 10 hr); earth is then ground to 200 mesh. Evaluation is by comparing bleaching power on a standard mineral oil (" Pebron 6 ") with that of a standard earth. Treatments are at 60° to 80° C, using 3, 5, or 10% earth, agitation 500 to 600 r.p.m., contact 10 min (vegetable oil) or 20 min (mineral oil). Colour measurement of oils, colour stability to temp, and merit rating of clays are considered. Evaluation may also be made on basis of measurement of removal of undesirable components as followed by changes in inter-facial tension. The second part of the paper deals with alkaline activation of clays. Ion-exchange properties are considered, and particular attention is paid to rheological behaviour of clay suspensions, and their colloidal nature. Procedures for evaluating shear resistance and swelling power are given. Sixty-six references. E. B.

1882. Investigation of optimum operating conditions for filter-press dewaxing. Braun and Raguideau. *Bull. Ass. franç. Tech. Pétrole*, 1951 (87), 53–64.—Mathematical relationships are derived between the factors involved in cold filtration, hot filtration, and sweating. Table is given showing wax yields at each of four successive sweatings. Based on this and on expressions derived, method is described for determining optimum operating conditions, *i.e.*, duration of cold filtration, wax % in filtrate from hot filtration, cutting point between first and second sweats. Nomograms for such calculations are given; conditions when refrigerating capacity is limited are also considered. V. B.

Special Processes

1883. I.C.I. Wilton works to make petroleum-derived terylene. V. S. Swaminathan. *Oil Gas J.*, 7.6.51, **50** (5), 86.—A new synthetic fibre will be produced in a plant now under construction at Wilton, Gt. Britain, based on starting materials derived from petroleum by the reaction of terephthalic acid with ethylene glycol. Starting materials are ethylene and paraxylene. G. A. C.

1884. Synthesis of hydrocarbons and of oxygen-containing compounds from carbon monoxide and steam. H. Kölbel and F. Engelhardt. *Brennstoff Chem.*, 1951, **32**, 150.—Preliminary notification of a new synthesis according to the overall equation $3CO + H_2O \rightarrow (CH_2) + 2CO_2$. Process is favoured by high pressure, but can also be carried out at normal pressure. Optimum temp is 200° to 300° C. Highly active cat can give yield of 195 g hydrocarbon per m^3 CO (at N.T.P.) (theoretical yield, 208 g.). CH_4 formation is relatively small; formation of O-containing products is favoured by higher pressures. Technical gases rich in CO can be used in place of the pure material. V. B.

1885. Spent acid recovery offers solution for tight sulfur supply. F. J. Bartholomew. *Refiner*, 1951, **30** (4), 101–3.—The best useful sulphur deposits in the U.S. are being exhausted. The use of pyrites for H_2SO_4 manufacture will be more expensive, and it is suggested that the petroleum industry should increase its acid-recovery processes. A normal spent alkylation H_2SO_4-recovery plant employing SO_2 to SO_3 conversion is described in detail. R. W. G.

Metering and Control

1886. A non-mathematical discussion of flow measurement. J. B. McMahon. *Refiner*, 1951, **30** (1), 99–106.—A brief review is given of work done in the U.S.A. on flow measurement. A fairly simple discussion of the principles underlying the use of orifice plates and venturi meters is presented, together with some practical notes on their uses. The article is aimed at providing for process men an understanding of some of the apparently vague and involved flow-meter factors. R. W. G.

1887. Automatic control equipment used in the petroleum industry. H. Weiss. *Bull. Ass. franç. Tech. Pétrole*, 1951 (87), 4–14.—Introductory lecture outlining main development features in this field; diagrams showing principles of main types of air-operated controllers are reproduced. V. B.

1888. Control and regulating equipment in the petroleum industry. P. Lefèvre. *Bull. Ass. franç. Tech. Pétrole*, 1951 (87), 15–26.—A study of the increasing complexity of control equipment (*e.g.*, in dist units, boiler installations) resulting in a decrease in operating staff, smoother operation, and an increasing reliance on instrument readings alone in processes handling tangible materials (*e.g.*, fluids); analogous to the reliance placed on instruments in electrical engineering, where the only indication of the flow of an intangible medium (*i.e.*, electricity) is given by such readings. V. B.

1889. Centralized control in industries furthered by monitoring systems. E. J. Grace. *Refiner*, 1951, **30** (1), 107–10.—An illustrated discussion of the use of high-speed monitoring systems in measurement and control of industrial processes. A combination of these systems with the recent introduction of the graphic panel offers a means for providing a convenient and compact information and control centre that is constantly under the supervision of an operator. R. W. G.

1890. Tuning automotive control systems. J. L. Serrill and L. E. Jewett. *Refiner*, 1951, **30** (1), 89–94.—A systematic method of adjusting modern automotive control instruments is discussed. Curves are presented to assist with the analysis of mal-adjustments. Improved automotive control can lead to improved plant capabilities —as with the temp control in cracking. Too high a temp leads to excessive coking, whereas a low temp affects the conversion, thus accurate temp is desirable. R. W. G.

1891. Unusual control valve practices in a petroleum refinery. C. W. Bates and J. R. Martin. *Refiner*, 1951, **30** (1), 111–18.—Several control-valve problems encountered at the Baytown refinery of Humble Oil & Blending Co., are discussed. Amongst these problems are: sulphur deposits on control valves, pilot-plant valves, butterfly-type valves (for fluidized solid), and control valves by-pass manifold. R. W. G.

1892. Control of gas compressor plants. H. C. Givens. *Refiner*, 1951, **30** (1), 95–8.—Deals primarily with the controls of an entire gas-engine-driven reciprocating compression plant, but most of the ideas advanced are also applicable to steam-driven reciprocating machines. R. W. G.

PRODUCTS

Chemistry and Physics

1893. Sur la flamme froide de l'ether. L. Ouellet and C. Ouellet. *Canad. J. Chem.*, 1951, **29**, 76.—Apparatus described which is used to study light emission of a cool flame of ether. Influential factors such as concentration of reactants, temp, and dia of the reaction vessel have been studied. The effects of these variables are fully discussed. A. D.

1894. The mercury photosensitized reactions of benzene at high temperatures. E. J. Y. Scott and E. W. R. Steacie. *Canad. J. Chem.*, 1951, **29**, 233.—No reaction found to occur at room temp, but at higher temp reaction takes place, the main product being diphenyl. Evidence is obtained that the activated molecular mechanism occurs. The secondary reactions are complex, and a detailed mechanism is not obtained, but the probable main reaction steps have been pointed out. A. D.

1895. The reaction of H and D atoms with cyclic and paraffinic hydrocarbons. H. I. Schiff and E. W. R. Steacie. *Canad. J. Chem.*, 1951, **29**, 1.—Reactions of these atoms, produced by the discharge method, with benzene, four *cyclo*alkanes, and corresponding

normal paraffins were investigated at room temp, major product being methane in each case. On the basis of hydrocarbon disappearance and exchange collision yields were calculated. Of the *cyclo*alkanes *cyclo*propane is most inert and *cyclo*pentane most reactive. No exchange observed between D atoms and either *cyclo*propane or *cyclo*butane, but considerable exchange takes place with benzene, *cyclo*pentane, and *cyclo*hexane. In normal paraffins, degree of exchange increases with increasing mol. wt. Concluded that initial reaction is

$$H + RH \longrightarrow R + H_2$$

responsible for products formed in these reactions, but other primary processes may be responsible for the exchange of these hydrocarbons. A. D.

1896. Thermochemistry for the petrochemical industry. Pt XV. Curve fixing: a method for minimum percentage deviation. K. A. Kobe and C. W. Arnold. *Refiner*, 1951, **30** (4), 123–4. (See Abs. Nos. 787, 2196–205, 2499, 1950; 436, 1443, 1951.)—Previous articles of the series presented thermochemical data on various hydrocarbons. This article presents a method of fitting an algebraic curve to experimental data. The method is direct and the possibility of large percentage deviations minimized. As an example, this method is applied to a set of values for heat capacities of *cyclo*hexane. The methods of "selected points," "averages," and "least squares" are briefly mentioned. R. W. G.

1897. Convection currents in porous media. III. Extended theory of the critical gradient. F. T. Rogers, Jr., and H. L. Morrison. *J. appl. Phys.*, 1950, **21**, 1177–81.—The theory of the critical gradient for onset of the thermal convection in a fluid entrapped in a porous medium is extended to allow for the exponential dependence of visc upon temp and for non-linear vertical temp—distributions which characterize transients in heat flow. Although there are approximations in the mathematical theory, the extended theory agrees well with experimental data, as the simple theory does not in certain instances.

New experimental data are reported for the critical gradient, obtained largely with silicone fluids in unconsolidated sands. N. T.

1898. Flocculation in suspensions of large particles. C. E. Hubley, H. A. Robertson, and S. G. Mason. *Canad. J. Res.*, Dec. 1950, **28B**, 770–88.—A new method is described of studying flocculation in suspensions of large particles based upon an assumed mechanism of particle aggregation by shear-induced inter-particle collisions, and representative results obtained with cellulose fibre suspensions are presented. The method consists in subjecting the suspension to a reproducible condition of shear motion and analysing electronically the fluctuations in optical transmission coefficients. From a statistical study, a flocculation index is proposed which is a function of both the number and size of aggregates and which decreases with increasing rate of shear and decreasing fibre length. Effects of concentration and of additives were investigated. Alternative experimental techniques are outlined, and the limitations of the present method are shown. N. T.

Analysis and Testing

1899. Accelerated ageing test limitations. C. A. Duval. *Refiner*, 1951, **30** (2), 85–6.—Accelerated ageing tests are criticized in that they may not always give consistent results. Data are presented showing that certain oils may be relatively more stable at 212° F than at 100° F. This is indicated more markedly with inhibited oils. However, it is pointed out that accelerated ageing tests are a necessity owing to the undesirability of waiting, possibly for years, for operating conditions to show their effects. R. W. G.

1900. Future holds great promise for continuous analyzers. B. W. Thomas. *Refiner* 1951, **30** (2), 81–4 (*Fifth Ann. Symp. on Instrumentation for Process Industries, Texas A. & M. College, Oct.* 1950).—Various continuous analysers, employing physical means, and their use in industry are discussed. Those specifically described are infra-red

analysers, densitometers, dielectric constant meters, and refractometers (using transmitted or reflected light). Applications referred to are, respectively: (1) determination of ethylene in ethane and methane, *iso*butane in normal butane and *iso*pentane, and ethane or *iso*butane in LPG propane; (2) recording and control of butadiene–styrene ratios in synthetic-rubber plants, etc.; (3) measurement of water, alcohol, or phenols in hydrocarbons; (4) estimation of vinyl and vinylidene chlorides, styrene in ethylene and butadiene, naphthenes in paraffins, aromatics in both paraffins and naphthenes, etc. R. W. G.

1901. Carbon dioxide and oxygen in natural gases. A. C. Lyles and R. A. Montague. *Refiner*, 1951, **30** (4), 93–4 (*California Natural Gasoline Assn Bull., TS*—501).—A tentative standard procedure for determining CO_2 and O_2 in natural gas is presented. It is similar to the Orsat method. Details of apparatus given include a permanent laboratory set-up claimed to be accurate to $\pm 0{\cdot}1\%$ of burette vol and a more robust field or plant unit claimed to be accurate to $\pm 0{\cdot}2\%$ of burette vol. Potassium hydroxide and chromous chloride solutions are used to absorb CO_2 and O_2 respectively in that order. R. W. G.

1902. A modified iodine pentoxide method for the determination of carbon monoxide. G. A. Grant, M. Katz, and R. L. Haines. *Canad. J. Tech.*, 1951, **29**, 43.—High concentrations of carbon monoxide can be measured satisfactorily, but while various methods have been derived for the determination of this gas in concentrations ranging from 0·01 to 0·001%, no one method is satisfactory on small samples of gas (500 ml). Improvements are described in the iodine pentoxide method. Refinements are made in the design of the apparatus, and the liberated iodine is estimated spectrophotometrically instead of by titration. The improved apparatus and methods are demonstrated by estimating the carbon monoxide concentration in ten mixtures of carbon monoxide and nitrogen ranging in concentration from 0·09 to 0·007% carbon monoxide. A. D.

1903. Correlation of oil-testing machines with practice. R. Halder. *Erdöl u. Kohle*, 1951, **4**, 180–6.—Overall factors governing choice and properties of lub oils are discussed and comparisons made of lubrication effects in moving parts of machinery under different conditions of oil visc and friction. Difficulties of relating tests with practice are described, and the importance of testing at operating temp stressed. Materials of construction in test machines are compared with practice, and effects of corrosion and erosion discussed with reference to the use of fresh test pieces for each test in order that limiting conditions may be achieved. Comprehensive survey is made of tests performed on various machines on one straight oil and the same oil doped with three additives, two phosphorus-containing compounds and one of unknown composition. Effect of additives is marked only in case of wear-tester (cf. Abs. No. 1926, 1950); in practice use of P-containing additives greatly reduced wear in Pb-bronze bearings of I.C. engines. Although individual tests can give misleading results a general correlation is shown to exist between the overall results obtained on various types of test equipment, and a method is developed comparing such results graphically and thus deducing properties of a lubricant under working conditions. P. V.

1904. Precise fractional distillation analysis. Theoretical investigation of low-temperature fractional analysis apparatus and methods. W. J. Podbielniak. *Refiner*, 1951, **30** (4), 85–92 (*Pet. Div., Amer. Chem. Soc., Atlantic City*).—A theoretical consideration of the fractionation of light hydrocarbons is presented, giving useful charts including relations between min reflux ratio and plates for different relative volatilities and product purities. The considerations are used in operating laboratory columns which are essential for the control of light-hydrocarbon-processing plants. This work has been reported in the literature before, but is published again on the assumption that it will be useful to plant personnel. R. W. G.

1905. Low-temperature fractional analysis of liquids and gases. Pt II. Fundamentals of operating technique. Causes and cure of errors. W. J. Podbielniak. *Refiner*, 1951, **30** (5), 145–55 (*Natural Gasoline Ass. America, Tulsa*). (See preceding abstract.)

—The previous article presented a theoretical investigation of low-temp fractional analysis apparatus and methods. This article supplements the earlier one with a treatment of the fundamentals of analysis apparatus, operating technique and of the causes and cure of errors. (Author's abstract.) R. W. G.

1906. A double beam recording infra-red spectrometer. K. S. Tetlow, J. McAuslan, K. J. Brimley, and W. C. Price. *J. Sci. Instrum.*, 1951, **28**, 161.—A description is given of this instrument, which uses a newly designed servo system and monochromator, the latter being compact and made from cheap and readily available components. Typical spectrograms illustrate instrument performance. C. N. T.

1907. Estimating pour points of petroleum distillate blends. E. B. Reid and H. I. Allen. *Refiner*, 1951, **30** (5), 93–5.—A method is presented for estimating pour points of blends from a knowledge of the pour points and ASTM 50% temp of the components of the blends. Any number of components may be considered, and it is claimed that an accuracy within the reproducibility of the pour-point test is obtained. The necessary data for obtaining the pour-point blending indices are given in a single graph. Only simple volumetric weighted arithmetic averages are used. This calculation method was developed through a thermodynamic consideration of the principles involved. R. W. G.

1908. A field method for the determination of halogenated hydrocarbons. P. E. Braid and K. Kay. *Canad. J. Tech.*, 1951, **29**, 159.—Describes a small portable field instrument for the estimation of methyl bromide in air. By means of a squeeze bulb a measured amount of test air is forced through a reaction tube containing a mixture of iodine pentoxide and fuming sulphuric acid, the free bromine so liberated being brought into contact with a fluorescein-treated test paper to produce a colour reaction. A colour card is provided to indicate concentration of methyl bromide from 0 to 150 p.p.m. by volume in air. Trichlorethylene, carbon tetrachloride, and higher concentrations of methyl bromide are determined by modifying the instrument and method. A. D.

1909. Instrument development at Thornton Research Centre. J. W. Drinkwater. *I.P. Review*, 1951, **5**, 97.—Shell's Thornton Research Centre deals with applicational problems arising from the use of petroleum products in areas outside the U.S.A. Thus the work is a mixture of chemistry, physics, and engineering. It includes an Applied Physics Section to develop and produce non-standard apparatus for the study of specialized problems. Whenever possible a "bread-board" model of the apparatus is used in the laboratory before the apparatus is provided in its finished form. This helps to find and to correct unexpected snags. Details are given of examples of this type of work relating to a four-channel cathode-ray oscilloscope, an engine programme-controller for testing the performance of fuels and lubricants for automotive engines, combustion problems of bunsen flames, a duel A.C. titrometer, an electronic engine-indicator, and oxygen-absorption testing. R. H.

1910. Micro-determination of sulphide sulphur (corrosive sulphur) in mineral oil. M. Zürcher and J. Lüder. *Bull. Ass. suisse Electr.*, 1951, **42**, 82–6; *Chim. et Ind.*, 1951, **65**, 668.—120 ml oil are contacted with metallic Ag in N_2 atm at 110° C. Silver sulphide formed on metal by contact with oil is reduced by H_2 at 450° C, the H_2S thus formed being determined iodometrically. S concn of 10^{-5}% can be measured. V. B.

1911. Oscillating friction machine for testing lubricants. H. L. Hendricks and J. D. Smith. *Institute Spokesman*, Jan. 1951, **14** (10), 7–17.—Frequent and early failures in the new individual front-wheel suspension systems of popular makes of automobiles are attributed, among other causes, to oscillating motion and chassis-lubricant breakdown. The necessity for the careful testing of lubricants is stressed, and details are presented of a new machine developed for this purpose. The equipment, which consists essentially of a sleeve-bearing system with means for loading and temp maintenance, is described in detail, together with test proceedure. Test results are tabulated, their interpretation is discussed, and the interdependence of grease properties in performance is considered, together with the possibilities of accelerated tests. J. G. H.

1912. Small electron microscope. J. H. Reisner and E. G. Dornfield. *J. appl. Phys.*, 1950, **21** 1131–40.—A new electron microscope with a resolution of 100 Å of greatly simplified design has been constructed. Magnetic lenses are energized by permanent magnets, yet it is small enough to be mounted on a desk. An accelerating voltage of 50 kV is varied over a narrow range to provide a means of focus. Images may be photographed on a 2 × 2 plate which can be changed in a minute; specimens are changed in fifteen seconds. Depending on pole pieces used, magnifications of 1500, 3000, and 6000 are possible. The instrument is free of X-radiation, is completely interlocked, and provided with a shorting device for protection from high voltages.
N. T.

Crude Oil

1913. Evaluation of Lacq crude. A. Giraud, A. Guillemin, and M. Lafitte. *Rev. Inst. franç. Pétrole*, 1951, **5**, 137–44.—Lacq field now produces 600 m³/day. Crude from well Lacq IV, having characteristics similar to those previously reported (see Abs. No. 2248, 1950), was fractionated (atm/vac) in pilot plant. Tests on fifty-four 1% cuts are tabulated, and properties and yields of gasoline, kerosine, and gas oil blends given. Gasoline O.N. (Motor) decreases (61 to 48) as yield and E.P. are raised from 9 to 16% and 149° to 222° C respectively. Effect of butane addition on gasoline was examined, and curves are also given showing relationship between yield and kerosine E.P. and S content of gas oil. M.E.K.–benzene–toluene dewaxing and furfural extraction of spindle oil fraction gave product of 72 V.I. and 2% S. 42% vac residue had d_{15} 1·052, m.p. (R. & B.) 41° C, pen 24, asphaltenes (hexane-insolubles) 26·6%, S 6·9%.
V. B.

1914. Experiments on the measurement of the viscosity of Emsland crude. S. Lechler. *Erdöl u. Kohle*, 1951, **4**, 286–7.—A qual account of visc measurements which indicate that Emsland crude exhibits thixotropy and structure-visc (non-Newtonian flow). The oil also shows sedimentation (*i.e.*, on standing a sample of gas-saturated crude shows a higher visc at the bottom of the sample than at the top). The crude is sensitive to oxidn which causes a marked visc rise. Microscopic examination shows that effects such as thixotropy and non-Newtonian flow are connected with wax crystallization.
V. B.

1915. Resins of Nienhagen crude. G. R. Schultze, J. Moos, and H. Witthuhn. *Erdöl u. Kohle*, 1951, **4**, 282–6.—Field of petroleum resins is briefly reviewed (thirty-six references). Crudes from N.W. Germany have resin content 9 to 27%. Fuller tests were done on Nienhagen crude (neut No. 0·5, S 0·8%, resin 10%) topped, freed from asphaltene by pptn with petroleum ether (30° to 50° C). Solution of crude in latter was passed through Al_2O_3 column, after de-oiling with petroleum ether, adsorbent was eluted with trichlorethylene to give light resin (m.p. 40° C, Br. No. 65, mean mol. wt. 400) and with ethyl ether to give heavy resins (m.p. 70° C, Br. No. 75, mean mol. wt. 650). Elementary analysis showed former to be essentially hydrocarbons (including six-membered rings), whilst latter were compounds containing, H,C,O. Petroleum resins give red colour with phloroglucinol and fuming HCl. Oxidn and cracking reactions of the resins were investigated and deductions made therefrom concerning their structure. Cracking of resins yields, *inter alia*, solid cyclic hydrocarbons, hence cracking procedures for paraffin-wax determination in crude and bitumen are unreliable.
V. B.

1916. Growing demand is seen for helium. R. A. Cattell and H. P. Wheeler, Jr. *Refiner*, 1951, **30** (3), 91–4.—Helium is being recovered in the U.S. from natural gas used for fuel, by liquefying nitrogen and hydrocarbons after removal of water vapour and carbon dioxide. The purchase price of helium from the Bureau of Mines is 1½ cents (U.S.) per cu. ft. Reference is made to the unusual physical properties of helium, and the several producing plants are discussed. By removing helium from fuel gas no depletion of state reserves actually takes place.
R. W. G.

Gas

1917. Mechanical sampling of fuel gases. E. A. McGlashen and J. S. Powell. *Oil Gas J.*, 31.5.51, **50** (4), 90.—Demand for more accurate measurement of physical properties of gas flowing through transmission and distribution systems has led to design of equipment for collecting a sample of gas in a portable container by mechanical means.

Apparatus consists of a gas-pressure regulator and spring-driven mechanism for the adjustment of the regulator screw at a constant rate.

The equipment is fully described and operating procedure given in detail.

The apparatus has been in continuous use at a gas-sampling location, by the Southern California Gas Co., and has given satisfaction. Figures show regulator system, arrangement of gas-control apparatus, and typical gas-pressure chart.

G. A. C.

Engine Fuels

1918. Trends in use of aviation fuel. M. Van Winkle. *Refiner*, 1951, **30** (5), 89–92.—A summary discussion of aviation fuels is presented, giving their compositions and availability. Since the second world war, the demand for aviation gasoline has declined and its specifications become more stringent. It demands: (*a*) improved anti-knock or performance quality; (*b*) a lower and narrower boiling range; (*c*) a lower sulphur content, and (*d*) a high lead susceptibility, requiring less T.E.L. per gal for the same performance quality. Various sources of aviation gasoline blending stock are discussed, including aviation naphtha, *iso*pentane, *neo*hexane, cumene, triptane, alkylate, and hydrogenated polymer. Turbo-jet fuels are also considered—these being more easily processed. Figures for U.S. aviation gasoline production over the years 1940–50 are given, and thirty-two references made to the literature. R. W. G.

1919. Vapour-phase production of dimethylaniline. T. H. Evans and A. N. Bourns. *Canad. J. Tech.*, 1951, **29**, 1.—The interest shown towards the end of the war in the anti-knock effectiveness of aromatic amines in aviation fuels caused this work to be undertaken. The object was the development of a process whereby dimethylaniline could be produced continuously by the vapour-phase reaction of methanol and aniline over solid dehydration catalyst.

This reaction and also the formation of dimethyl aniline by vapour-phase reaction of aniline and dimethyl ether were fully investigated. In both cases activated alumina was found to be the most efficient catalyst. The reaction with dimethyl ether was preferred, since higher yields of purer product were obtained at lower temp. Aniline reacted with five mol. dimethyl ether over Alcoa activated alumina. At 275° to 285° C a quantitative yield of dimethylaniline of 99% purity was obtained. Other catalysts tried were Harshaw activated alumina, Porocel (an activated bauxite) and Unigel, a Harshaw alumina gel. A full description is given of a pilot plant which was constructed and operated continuously for the production of 4 to 5 lb/hr dimethyl aniline. On the basis of yields, purity, catalyst life, etc., it is concluded that this process should be suitable for the large-scale manufacture of dimethyl aniline. A. D.

1920. Evaluation of motor fuels for high-compression engines. M. M. Roensch and J. C. Hughes. *S.A.E. Trans.*, Jan. 1951, **5** (1), 71–80.—Details are presented of a series of tests on a G. M. high-compression test engine arranged so as to permit operation at C.R. from 6/1 to 12/1. Over this range the behaviour of a variety of experimental and commercial gasolines was studied. It was found that increasing the C.R. from 6/1 to 12/1 gave b.h.p. gains up to 26% with equivalent gains in fuel economy. J. G. H.

1921. Gasoline consumption and travel time of trucks. C. Saal. *S.A.E. Trans.*, Jan. 1951, **5** (1), 94–110.—The value of data on gasoline consumption and travel time of commercial vehicles are emphasized, and results are reported of a co-operative investigation between Government departments and trade associations. The routes, vehicles, and procedures used are detailed, and the conclusions reached include that,

in any highway section, gasoline consumption and travel time vary directly with the rate of rise and fall, regardless of the length of the section; gasoline consumption is definitely related to the gross weight, and the travel time to the weight/power ratio. Results of the investigation can be applied to paved highways and gasoline-powered commercial vehicles in any part of the country where the average altitude of the highway section does not exceed 3000 ft. J. G. H.

1922. Study of mixture distribution. R. W. Donahue and R. H. Kent. *S.A.E. Trans.*, Oct. 1950, **4** (4), 546–58, 570.—The background of mixture distribution problems is briefly outlined, and data are presented on an investigation using a single 1946 six-cyl engine. The apparatus and procedure used are detailed, and results obtained indicate that such design features as the position of the choke valve and its mounting and any other obstructions in the air passage can alter the quantitative mixture distribution patterns more than reasonably large changes in engine speed, mixture temp, or fuel volatility; that the lightest components of fuels are unevenly distributed as well as the heavier materials; that the anti-knock quality of the fuel reaching each cylinder depends to a large extent on the variation in octane quality with boiling range of the original fuel, and this quality can be controlled to a considerable extent by maintaining even quality between the light and heavy fractions of the fuel. J. G. H.

1923. Effects of sulfur in motor gasoline on engine operation. R. E. Jeffrey, J. B. Duckworth, and E. J. Gay. *S.A.E. Trans.*, Jan., 1951, **5** (1), 111–22.—Results of a CRC investigation of the effects of sulphur in gasoline on engine operation are summarized. Sixty-two vehicles completed tests at seven test sites to compare the effect of two gasoline sulphur levels 0·10 and 0·25%, on engine wear and cleanliness. The data indicate that the influence of high sulphur content may vary from a pronounced effect on cylinder and ring wear at some operating conditions to no effect under continuous operating conditions at normal engine temp. Discrepancies in the procedure adopted are noted for correction in future investigations. J. G. H.

1924. The rise and fall of the petrol bus. A. T. Wilford. *I.P. Review*, 1951, **5**, 147.—Although buses of various kinds were in operation in Britain from 1829 onwards, it was not until 1900 that petrol-driven buses were tried in London. Intensive development took place from 1900 to 1908, and many varied designs were seen. However, frequent breakdowns took place, and in 1908 there were still only just over 1000 of this kind of vehicle. 1910 saw an increase in activity, and from then onwards the growth was steady, and the types varied. A six-cylinder engine with overhead camshaft was used in a six-wheel three-axle type bus in 1929, the two-axle model following in 1930. This engine was produced in 37- and 45-h.p. sizes with a C.R. of 5 to 1, and a max useful speed of 2500 r.p.m. C.I. engines were tried in 1929, and soon replaced petrol-engine types to a great extent. The last petrol-bus was withdrawn from London in 1950. Diesel engines have definite advantages over petrol engines for buses. There is less fire risk, the load–speed characteristics make it particularly suitable for heavy traffic conditions, and a higher thermal efficiency (34% compared with 23%) gives a substantial economy in fuel consumption. R. H.

1925. Combustion and fuel particles. D. B. Spalding. *Fuel*, June 1951, **30** (6), 121–30.—The combustion theory described in an earlier paper is applied to the combustion of small particles of oil and carbon under conditions in which convection is absent, and is extended to cases of variation of thermal conductivity with temp and dissociation of the gases. The combustion of a droplet of kerosine, a comparison of solid and liquid fuel combustion, and the effect of engine conditions, covering limited oxygen supply, uneven distribution, turbulence, swirl, injection, and ignition, are described. The combustion rates of fuel particles are calculated and compared with the time available for combustion in diesel and gas-turbine engines. W. H. C.

1926. Elimination of combustion knock—Texaco combustion process. E. M. Barber, B. Reynolds, and W. T. Tierney. *S.A.E. Trans.*, Jan. 1951, **5** (1), 26–42.—The importance of maximum fuel utilization is emphasized, and the objects and methods of the Texaco combustion process (T.C.P.) are described. The basic concept of T.C.P.

is to make the residence time of combustible mixture so brief that spontaneous ignition and knock will not occur at engine pressures and temp. In this manner it is claimed that compression ratio may be increased without regard for the O.N. of the fuel, supercharging may be employed with high compression, if so desired, without regard for the O.N. of the fuel, lean mixtures may be burnt, resulting in a favourable part-load economy, and fuels can be utilized having no O.N. specification and broad boiling range. J. G. H.

1927. Precombustion reactions in a motored engine. D. L. Pastell. *S.A.E. Trans.*, Oct. 1950, **4** (4), 571–87.—Present paper is the first report of a general project on end-gas reaction studies, having as its broad objective the development of new data on chemical factors causing knock. Previous methods of attacking the problem of knock are noted, and the advantages of using a motored engine are enumerated. The equipment and procedure used are indicated, and it is concluded from the results obtained that precombustion reactions are affected by chemical and physical variables in much the same manner as these effect knock. It has also been found that the temp and pressures required for trace knock in a fired engine are approx the same as those required for auto-ignition in a motored engine operated at the same speed, indicating that precombustion reactions leading to auto-ignition are probably the same as those leading to knock in a fired engine. J. G. H.

1928. Precombustion reaction in the spark-ignition engine. E. R. Retailliau, H. A. Ricardo, and M. C. K. Jones. *S.A.E. Trans.*, July 1950, **4** (3), 438–54.—Previous studies have indicated that gasoline undergoes charges during the compression period prior to the point where spark ignition would normally occur, these charges being known as "pre-reactions." Paper is concerned primarily with the effect of engine-operating variables on the pre-reactions which occur when employing different hydrocarbon types as fuels. The nature and effects of pre-reactions are discussed, and the equipment and procedures used are outlined. Engine-operating variables considered include intake temp of air–fuel mixture, temp, heat capacity, and amount of unscavenged exhaust gas present from previous cycle, engine-jacket temp, and C.R. Under the conditions investigated benzene and aromatic-type feeds appear to resist pre-reaction, while paraffinic-type hydrocarbons tend to pre-react; this tendency can be minimized by the addition of T.E.L. J. G. H.

1929. Cylinder performance—compression ratio and mechanical octane-number effects. S. D. Heron and A. E. Felt. *S.A.E. Trans.*, Oct. 1950, **4** (4), 455–99.—Data obtained with a cylinder of 3¼ inch bore by 3¼ inch stroke are discussed, obtained with compression ratios of from 5·7 to 15 to 1 with four types of compression chambers comprising standard and shrouded intake valves with flat top and squish pistons. The investigation includes determination of fuel economy at 1200 and 3000 r.p.m., with particular emphasis on part-load economy. In order to facilitate comparisons over a wide range of C.R., fuel-economy data are presented in terms of relative thermal efficiency; knocking data in terms of air density. It is shown that over a considerable C.R. range, the knock-limited combustion-chamber air density on *iso*-octane or 80 octane–70 heptane is independent of C.R. The effect of turbulence in improving part load economy and on engine severity is also discussed. J. G. H.

1930. Purification of diesel-engine fuels in relation to cylinder-liner wear, with special reference to the employment of boiler oil in motor ships. J. Lamb. *Motor Ship*, May 1951, **32** (374), 55.—Experience with Anglo-Saxon tankers operating on ordinary commercial grades of fuel, of visc varying from 100 to 3500 sec Red. I at 100° F, has proved that engines require no more attention than when operating on diesel oil and that wear rates of cylinder liners and piston rings are no greater when burning the cheaper grades of fuel. Correct treatment of fuel and operation of engine is, however, essential when using viscous fuels, and fundamentals of fuel purification are outlined here. Insoluble ash must be completely removed, either by settling out or by centrifuging, and both processes are facilitated by heating. It is important that throughput of centrifugal separators should not be greater than that found necessary to extract whole of insoluble ash content. Extraction of waxy constituents should be avoided, and to this end purifying temp for relatively light fuels should be varied,

pre-heating sometimes being unnecessary. Injection temp requires to be between 160° and 200° F, and particular attention is required when starting an engine from cold or running at slow speeds for long periods in cold climates. Particulars of performance and cylinder-liner wear in two ships are included. U. M.

1931. The petrochemical engineer looks at rocket fuels. M. Sittig. *Refiner*, 1951, **30** (5), 114–16.—The requirements of rocket fuels are listed, and many fuels, monopropellants, and bipropellants, are indicated. Reference is made to the German second world war rocket fuels and to the operation of the V2, Additives, such as terpene hydrocarbons, may be used as combustion catalysts. The manufacture of several rocket fuels is discussed, including ammonia, hydrazine, aniline, nitric acid, nitromethane, furfuryl alcohol and tetrahydrofuran, methanol, methylal, ethanol, vinyl ethers, boron hydrides, and hydrogen peroxide. R. W. G.

Lubricants

1932. Hydrodynamic lubrication : general survey. D. G. Christopherson. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication, Pt* 1). *Symp. of British Rheologists' Club and Inst. Physics, Manchester*, June 29–July 1, 1950.—The classical work of Reynolds in 1886 is first reviewed, and the basic assumptions of lubrication theory are stated. Three or four of these are classed as "essential," *i.e.*, fundamental to progress, and the remainder as inessential but "simplifying." The properties of lubricant films are surveyed, the variables considered being temp and pressure.

An account is given of the properties of bearing metals, under the headings of surface roughness and general deformation. Finally, the question of the occurrence of negative pressures and cavitation is given attention. It is concluded that there is now a wider realization that wear reduction and longer life result from ensuring hydrodynamic lubrication conditions, and also that the knowledge of design and of lubricant selection and treatment is becoming more widespread.

J. A. Cole, in contributing to the discussion, referred to the question of film extent, which is important in calculating friction torque, and inquired as to the adequacy of various boundary-conditions in assessing bearing performance.

G. W. Nederbragt drew attention to the possible influence of the thermal conductivity of oils, since the ratio of the conductivities of paraffinic and naphthenic hydrocarbons might be as high as 1·7. The thermal conductivities of C_{16} and C_{24} *n*-paraffins are relatively high, and increase with mol. wt. For the naphthenes studied, the values are relatively low, and decrease with mol. wt., whilst the figures for castor oil are nearly three times that of a fluorocarbon oil. C. N. T.

1933. The boundary layer. G. I. Finch. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication, Pt* 2), p. 34.—The present state of knowledge of the liquid and solid boundary layer is reviewed. The function of the boundary layer is to replace the oxide-layer component of friction by one of still lower value. The structure of the oil-boundary layer is discussed, and the effects of heat and sliding on this structure, particularly on the orientation of the lubricant molecules. Solid lubricants of the layer lattice type depend for their functioning on a large ratio of surface area to particle mass, and having on their surfaces firmly adhering liquid or gaseous adsorbed layers. (Author's Abstract.) C. N. T.

1934. A study of boundary lubricant films by electron diffraction. J. W. Menter. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication, Pt* 2), p. 52.—The work described was on the orientation and structure of thin films of long-chain hydrocarbons on metal surfaces. The effect of temp was studied, the results being correlated with the frictional behaviour of lubricated sliding surfaces. It is concluded that good boundary lubrication occurs only where there is a closely packed solid film of oriented molecules on the surface.

In opening the discussion, K. H. R. Wright pointed out discrepancies between the author's results and disorientation points obtained elsewhere. F. R. Dean inquired about the uniformity of monolayers, and the author replying, mentioned three main differences in experimental technique which might explain the discrepancies, and cited evidence on the question of monolayer packing, mentioning that work was in progress on this problem. C. N. T.

1935. An introduction to boundary and extreme pressure lubrication. D. Clayton. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication, Pt* 2), p. 25.—After indicating the relationship of boundary and fluid film lubrication, and the importance of the former, reference is made to the explanations of boundary friction, in particular to the adhesion theory; the simplified explanation of the laws of friction in terms of this theory is given. Dealing with the influence of the main variables on friction values, velocity is first considered, together with the explanation of friction-induced vibrations in terms of the main type of friction/velocity curve; a simple, general statement on the effects of temp and surface roughness on friction cannot yet be given. Brief references are made to the high temp reached superficially at sliding contacts, and to electrical effects. In terms of these fundamental conceptions, a sketch is given of the mechanism of running-in, of wear (including notes on wear testing and anti-wear additives), of the behaviour of surfaces under severe conditions leading to scuffing (including the function of E.P. lubricants).

Finally, the value of solid lubricants is mentioned, and also the benefits that can be obtained by chemical pre-treatment of the solid surfaces. At each appropriate point of the survey reference is made to the other papers presented at the conference. (Author's Abstract.)

In the discussion, F. T. Barwell dealt with matters of definition, and suggested the possibility of a third state, termed "thin film lubrication." A hypothetical mechanism for the different types of friction–velocity curves was put forward for discussion.

H. Blok emphasized that in hypoid-gear operation, the contact temp is more critical than the contact pressure in regard to teeth scuffing, because of the different type of motion involved. Contact temp were appreciably higher than those in spiral bevel gears, but it would be hard to replace the misnomer "E.P. lubricants" by an alternative term.

E. D. Tingle mentioned evidence of surface-layer formation, and suggested that in boundary lubrication there might be several regions, depending on layer structure of the sliding surfaces and load. C. N. T.

1936. Scuffing temperatures of boundary lubricant films. T. B. Lane. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication, Pt* 2), p. 35.—The postulate of Blok that lubrication will fail when the temperature in the contact area reaches a value characteristic of the lubricant and sliding system is confirmed by experiments on the two-ball machine. It is shown that, for the range of mineral oils tested, the failure temperature increases with viscosity. (Author's Abstract.) C. N. T.

1937. Physical changes in rubbing surfaces on scuffing. D. Clayton and C. H. M. Jenkins. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication, Pt* 2), p. 69.—Cast-iron surfaces rubbing against steel develop a thin layer of "white constituent" when scuffing occurs or is near. The material is not positively identified, but it is suggested it has as basis two phases (1) cementite and (2) quenched high-carbon ferritic phase developed from austenite. With a four-ball apparatus, modified to take piston-ring segments, the white material was also obtained in some degree with mineral oil under milder conditions than those causing scuffing, but not with the castor oil, which proved better in preventing scuffing in running-in an aero-engine.

This war-time work was not completed, but the results are recorded to draw attention to the phenomenon, and may also act as pointers to further work needed to elucidate running-in effects and scuffing.

A. A. Milne, in opening the discussion, outlined various physical and geometrical effects which occur on scuffing, in addition to metallurgical changes.

J. E. Stringer asked whether the changes in the manufacture of cast iron or in the refining of mineral oils were more likely to explain why, in the last twenty to thirty years, scuffing troubles with cast-iron valve plates in hydraulic machinery appear to have become more pronounced.

The authors, replying, thought that improvements in cast-iron quality were much more likely to explain the differences, although oil-refining developments may have had some effect. C. N. T.

1938. Surface welding and the extreme pressure lubricant. G. I. Finch and R. T. Spurr. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication, Pt* 2), p. 79.—Surface oxide films prevent inter-metallic welding when two surfaces are brought into contact,

but may be ruptured by sliding or by applying sufficient pressure to cause plastic flow at the interface. The ease with which surfaces weld on sliding depends on their rigidity and on the surface film nature and thickness. E.P. additives react with the surface oxide, forming a compound more fusible than the metal. If local temps rise enough during sliding, the compound will melt, smear over the surface, and act as a lubricant.

A. W. Crook in the discussion referred to experiments showing that the strength of the metallic junctions formed when two surfaces are brought together depends on the time available for their formation.

J. S. McFarlane outlined the results of his adhesion experiments with D. Tabor, from which he concluded that the mechanical and surface properties of the metals are also of importance. C. N. T.

1939. The spreading behaviour and lubricating properties of mixed liquids. E. B. Bielak, E. W. J. Mardles, and E. D. Tingle. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication, Pt* 2), p. 58.—Measurements of the rate of radial flow of liquids and binary mixtures over or between polished surfaces have been correlated with the corresponding friction results. The results have been analysed, and are presented graphically. It is concluded that changes in the molecular association of the liquids on admixture appear to account for the observed behaviours. C. N. T.

1940. The electrostatic component of the force of friction. R. Schnurmann. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication, Pt* 2), p. 62.—Experimental evidence has been presented previously in support of the view that jerky or "stick–slip" motion of a frictional contact operated at a uniform rate of propulsion was of electrostatic origin. The electrification of frictional contacts is shown by discharge currents made visible on a C.R.O. screen. With a metallic contact against a rotating steel disc, rapid oscillations are found, whose amplitude is independent of load and speed (in the region of 400 cm/sec linear motion), but can be increased by adding a dielectric. The oscillations can be made to disappear under conditions where static charges cannot be held. Similar effects are observed using a rotating glass disc, when luminous spots are observed below 400° C at the contact area.

F. T. Barwell, in the discussion, asked whether the results were corrected for the hot-junction thermal capacity; Dr Courtney-Pratt disputed the author's theory that electrostatic forces formed a significant part of the forces of friction, and cited evidence in support of his contrary view.

A. J. Chisholm asked about the "hot-spot" contact temp, whilst J. E. Stringer suggested resolving the question of whether or not the effects were of frictional or thermo-electic origin by practical experiment. The author replied to the various points in some detail. C. N. T.

1941. Adsorption of lubricant films: a study by radioactive tracers. A. C. Moore. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication, Pt* 2), p. 54.—Chemical reaction between radioactive metal surfaces and adsorbed monolayers of stearic acid, ethyl stearate, and octadecyl alcohol was investigated. When reaction occurs, good lubricating properties are shown at temp above the bulk melting point up to the soap-softening point. If no reaction occurs, lubrication is found only below the bulk melting point of the lubricant. When there is a reaction, it is not restricted to monolayer formation of soap, and if enough material is present, thick films are built up.

W. Hirst, in the discussion, referred to experiments by Lancaster, using the powder/solution technique (*i.e.*, reaction between metal oxides and stearic acid in benzene). The results depended on the extent to which the materials were dried before use. With Cu and Zn, and their oxides, the amount of water present affected the reaction rate, and it was thought that under dry conditions, stearic acid is physically adsorbed, and that chemical reaction does not begin until a certain minimum concentration of water diffuses to the surface. The evidence suggests now that this hypothesis is correct. Once started the reaction between stearic acid and metal oxides produces water, the effect of which is presumably then of less importance.

A. J. Chisholm referred to work on the behaviour of fluids used in machining metals, where a low-shear-strength substance may be formed at the interface by chemical reaction. C. N. T.

1942. Lubrication with materials in the solid state. F. T. Barwell and A. A. Milne. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication, Pt* 2), p. 39.—Coefficients of friction were measured between flat mild-steel surfaces loaded at an average intensity of 40 lb/in^2 at two relative velocities using a variety of solid lubricants. Only graphite, metallic soaps, and paraffin wax were effective lubricants, and the last was studied further under more practical conditions. Chemically treated mild steel was reciprocated against the untreated material with and without lubricant. A phosphated surface, lubricated with paraffin wax, was the most satisfactory. In these experiments the porous nature of the surface plus the rheological properties of the wax might govern the performance.

E. A. Smith, during the discussion, presented data which indicated that the coefficients of friction for graphite/oil films show a comparatively wide spread, whilst for graphite alone, the metal plays no part if fully covered. Values higher by 20 to 50% are obtained for mineral oil, which may therefore adsorb more readily on graphited than on plain metal. Reference was made to industrial applications of graphite in oil.

In answer to A. F. H. Ward, the author explained that he had used powdered lubricants. The results did not indicate that graphite would behave better in oil than polar or E.P. additives. C. N. T.

1943. The lubrication of aero gas turbines. J. G. Dawson. *Voordr. Koninkljik Inst. Van Ingenieurs*, Jan. 1951, pp. 132–48.—Data obtained at the Thornton Aero Research Laboratory are given with reference to gas-turbine lubricating oils, and are discussed from the aspects of the basic requirements of (*a*) gears and (*b*) bearings, and the requirements of (*c*) high-temp and (*d*) low-temp, operation. Oils to Specification D.E.D. 2479 are used generally in propeller turbines, and D.E.D. 2480 and D.T.D. 44D. are used for pure jet engines. These are discussed as to their merits and defects, additives for their improvement with respect to gear loadings, visc, V.I., pumpability, etc. Future development of an oil for propeller turbines must be directed towards improving load-carrying capacity in gears and reducing the pour point and visc at low temp. In pure jet engines oils development must be towards an improvement in V.I. in order to retain the present characteristics at low temp and at the same time provide a more viscous oil at higher temp. No vast improvements in mineral oils can be foreseen, except in the use of E.P. additives. Any major advance must be sought in the field of synthetic oils. Synthetic oils are reviewed, and their outstanding characteristics discussed. Their cost, compared with straight mineral oils and with the total operating cost of a gas turbine, is outlined. W. H. C.

1944. Light engine oils for improved subzero operation. V. G. Raviolo. *S.A.E. Trans.*, Apr. 1950, **4** (2), 161–7, 286.—The necessity for an oil lighter than S.A.E. 10 for subzero operation is noted, and circumstances leading to the formation of CRC Group on Engine Oils lighter than S.A.E. 10 are outlined. Details are presented of the introduction and properties of 5W oils, and the test programme evolved is summarized. Points dealt with in some detail include visc, stability, oil consumption, cold-starting properties, and wear. In all cases 5W performance as compared with that of 10W is superior, with the exception of oil consumption, where an average increase of 30% is experienced with 5W oils. J. G. H.

1945. The lubrication of cotton and other textiles. E. Moss. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication*), p. 19.—The frictional properties of cotton are controlled by the layer of natural wax on the fibres. Those finishing processes which remove wax tend to increase friction between cotton and other surfaces. Introduction of other surface layers, as in dyeing, may also increase friction. The high friction of wax-free cotton is reduced by the addition of lubricants, whose effect within some classes of compound varies regularly with chemical constitution. The lubrication of regenerated celluloses is broadly similar. Low and uniform friction between textile and machine is very important in processes such as knitting and stitching. (Author's Abstract.)

A. Perelman, in the discussion, tried to reconcile the author's finding that the coeff of friction of cotton against steel increased with paraffin lubricant chain-length with some apparently contradictory Dutch results.

A. A. Milne drew attention to the complete reversal (in order of increasing friction)

between lubricants used in textile operations and the same ones used under boundary lubrication conditions with metallic surfaces, at low velocities and high pressures.

In his reply, the author did not agree that lubricants which are " good " for metallic surfaces are " bad " for textiles. C. N. T.

1946. The lubrication of metals by long chain organic compounds. K. V. Shooter. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication, Pt* 2), p. 49.—Previous work on boundary lubrication of metals by fatty acids has shown that for efficient lubrication, chemical reaction and soap formation must occur. More than an adsorbed soap monolayer on each of the sliding surfaces is required to produce a low coeff of friction and good lubrication.

In the discussion A. F. H. Ward drew attention to the delay in adsorption of long-chain fatty acids on to a water/solvent interface, which might be relevant to the present work. If a delay was detectable by friction measurements, it would be important where the film was broken locally by incipient seizure. Speed of adsorption might be the dominant factor in lubricant efficiency. The author, replying, said that his monolayers were formed in a few seconds, but noted that other workers had observed measurable delays. C. N. T.

1947. The effect of lubrication on friction, wear, and abrasion. P. Grodzinski. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication, Pt* 2), p. 86.—Lubrication of grinding surfaces is important in abrasive processes, and recent measurements by an electronic dynamometer of the coeff of friction (cast-iron disc/lubricated diamond surface) gave values for various oils generally below 0·08 without diamond powder and about 0·11 with the powder.

A. J. Chisholm, in the discussion, likened grinding to a cutting process, and gave an alternative interpretation of the author's experiments, based on this view, to which the author replied. C. N. T.

1948. Lubrication of rough steel surfaces by a series of metallic soaps. F. T. Barwell and A. A. Milne. *Brit. J. Appl Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication, Pt* 2), p. 44.—Friction experiments are described where metallic soaps and other materials were used to lubricate rough steel surfaces, under one set of surface conditions at various velocities and temp. An empirical relation $\log Fa \log V + K$ (where F = friction, V = velocity, and K = constant) was found to be obeyed closely by metallic soaps, but less well by methyl stearate and docosane. The melting point of the solid lubricants was very critical, higher temp giving a sudden friction increase, whilst for greasy soaps, friction rose continuously from 25° to 150° C. Friction was affected little by changing the metallic ion or acid radical, except where this altered the physical state. In the case of stearic acid, there was evidence of progressive reaction with the metal surface. Methyl stearate was a less effective lubricant than the soaps, whilst docosane was very poor. Microscopic examination showed that the solid lubricants were very effective in separating the surfaces, contact occurring only at intermittent points. Much greater contact occurred with melted lubricants, this contact being only at the peaks of surface irregularities, which were depressed by a relatively small amount. C. N. T.

1949. An investigation of surface damage using radioactive metals. E. Rabinowicz. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication, Pt* 2), p. 82.—In studies made using a radioactive metal hemisphere sliding over a non-radioactive metal surface, the amount and nature of the transferred material were examined autoradiographically. Metal transfer took place in the form of discrete fragments; well-lubricated surfaces gave at least 400 times less transfer, but on heating them, both the coeff of friction and the amount of metal transfer increased markedly at the m.p. of the surface film. Metal transfer, but not friction, was sensitive also to temperature changes below the m.p., presumably because of film softening. In some instances pick-up of large non-radioactive fragments by the slider prevented radioactive fragment transfer. C. N. T.

1950. The rheology of lubricant films. E. W. J. Mardles. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication*), p. 7.—The rate of radial spread of a pool of liquid

over polished horizontal surfaces or between optical flats, approaching or receding in a normal direction, has been measured for several lubricants and liquid mixtures. The quantitative results obtained have indicated divergences from normal hydrodynamic behaviour, and are specific to the kind of lubricant and of the material of the surface.

During the thinning of liquid films, rupturing with " pin-holing " has been observed, the stage of onset varying with the kind of surface and lubricant. Evidence of the " wringing " together of flats has been obtained from sliding results at low pressure and by the separation of totally immersed flats by normal pull with small forces. (Author's Abstract.) C. N. T.

1951. Viscosity of lubricating oils at high rates of shear. A. F. H. Ward, S. M. Neale, and N. F. Bilton. *Brit. J. Appl. Phys.*, 1951, *Supp. No.* 1 (*Physics of Lubrication*), p. 12.—Visc coeff of lubricating oils have been measured, using a high-speed rotational viscometer, with a very thin oil-film. Special attention was given to the estimation of the temp of the oil, so that the fall in visc caused by the rise in temp might be allowed for. It was then found that at high rates of shear the coeff of visc showed a decrease which was a function both of the rate of shear and of the temp. This could be interpreted in terms of a balance between the orientation produced by the velocity-gradient and the disorientation caused by the Brownian motion. A much more marked anomalous effect was shown when a V.I. improver was added to the oil. (Authors' Abstract.)

In the discussion, H. Blok made various comments and suggestions on the basis of published results, both theoretical and experimental. The authors' results seemed to prove beyond doubt that at least certain straight mineral oils can exhibit a real visc fall under rates of shear lower than the theoretical value. C. N. T.

Bitumen, Asphalt, and Tar

1952. A study of cold water separation of bitumen from Alberta bituminous sand on a pilot plant scale. L. E. Djingheuzian and T. E. Warren. *Canad. J. Tech.*, 1951, **29**, 170.—Method for this separation consists basically in adding an oil diluent, water, an alkaline reagent, and a swelling agent to bituminous sand, disintegrating the mixture in a pebble mill, diluting with water, separating the major part of the sand in a classifier, and settling out water and finely divided inorganic matter in thickeners. The diluent is distilled from oil product and recycled. This has been performed in a pilot plant having a capacity of 200 to 300 lb/hr of sand. Recovery of total oil usually 97·5% approx, and net bitumen recovery about 95%.

Effects of many variants have been investigated. A. D.

1953. Investigation of the adhesiveness of bitumen to stone under the action of water. E. Neumann. *Bitumen, Teere, Asphalte, Peche*, 1951, **4** (2), 89–93.—The problem of the adhesiveness of bitumen to stone in road construction is studied with special reference to the effect of water. For physical testing the Swedish Road Construction Institute, in its latest report No. 78, introduces a new apparatus as illustrated, in which the change by wt between bitumen and water at the surface of the stone is measured. Adhesion and surface-tension measurements show that the former depends on a combination of a definite binder with a definite stone. Heavy oils are superior to light oils, tar oils to the corresponding petrol oils. Four types of adhesion are discussed. Laboratory tests do not correlate well with road tests. Experiments carried out at Stuttgart Research Station with special attention to grain structure, filler content, and binder addition, especially to pore space, confirmed the detrimental influence of water, but by careful compounding of the masses and reducing the pore space to a min this unfavourable influence can be cut out. Experiments carried out by the American Bureau of Public Roads, which simulate the conditions of those of the Stuttgart Research Institution, are described. Their value lies in the proposals for the special treatment of the masses, in the choice of a suitable procedure for water storage, and in the ascertaining of the varying behaviour of different additions, fillers, and bitumen. The effect of water on the adhesiveness of the binder is important only for types of open construction. The pH of the water is important. R. T.

1954. Researches on the constitution of natural bitumens. VI. Extraction of bitumen by liquid sulphur dioxide. E. Mariani and R. Turriziani. *Boll. soc. Fac. Chim. ind. Bologna*, 1950, **8**, 98–100; *Chim. et Ind.*, 1951, **65**, 685–6.—Results of tests on the solubility of natural bitumens in liq SO_2. Extracts obtained are reddish very viscous oils. These have been fractionated into asphaltenes, oils, and resins. Liq SO_2 has preferential solubility for the oils, solvent power decreasing for resins and asphaltenes. C/H ratio of bitumen asphaltenes removed by SO_2 is less than that of asphaltenes dissolved by C_6H_6 and by other bitumen solvents (*e.g.*, CS_2, CCl_4). Extraction by SO_2 is accompanied by polymerization phenomena. V. B.

1955. Bitumen decomposition with the aid of selective solvents. Pt II. Practical application of the method. K. Krenkler *et al. Bitumen, Teere, Asphalte, Peche*, 1951, **4** (2), 85–8.—Selective solvents *n*-butanol, *n*-heptane, and *cyclo*hexane discussed in Pt I, **3**/51, are applied to the investigation of bitumens of different origin and production. Extraordinary variations in the composition of apparently similar bitumens are established. Such variations—important for the practical use and rating of bitumens—are not detected by conventional methods. The medium mol *n*-butanol insol resins bear adhesive properties and ductility but possess the disadvantage of temp sensitivity, lower stability, and brittleness. The high mol *n*-heptane insol gives the bitumen plasticity, low temp sensitivity, and the true characteristic properties of bitumen. The highest mol *cyclo*hexane insol constituents are no longer colloidally dissolved, but exist as micro-particles like finely divided fillers, and do not contribute to the binding properties of the bitumen. Compared with steam-distilled bitumens, blown bitumens undergo a transformation of the components whereby the medium mol resins are converted into high-mol material by polymerization and condensation, resulting in a change in properties due to a change in micelle orientation. The results of the investigation presented in diagrams show clearly the operation of the blowing process on the internal structure of bitumens. Further, the great differences between various series of blown bitumens which have the same apparent properties are indicated. R. T.

1956. Influence of water on the stability of composition roofing. A. W. Rick. *Bitumen, Teere, Asphalte, Peche*, 1951, **4** (2), 94–6.—Testing of composition roofing and protective measures against the effects of water are discussed. German specifications contain no proof test for the resistance of these roofings to water. The properties of composition roofings in relation to water were ascertained by the following tests: water absorption, strength in the wet condition of strewed compositions, extension in the moist condition, permeability to moisture. Occluded water has a more serious effect than saturation moisture. It renders the vital activities of fibre-destroying bacteria possible unless these are stopped by impregnating solution. Also during swelling it causes single threads to become loose. The third phenomenon, blister formation, is a source of perpetual trouble. The blisters burst on the surface and form funnels through which water enters. These conditions may be remedied by the application of hot or cold bitumen coatings. Only coatings of solvent medium have a lasting effect, according to the conditions, the results are still better if a sufficient quantity of heavy oil is available. The oil renews the pliability of the composition roofing and closes the pores. R. T.

Special Hydrocarbon Products

1957. Cutting oil Hö. G. Schulz. *Erdöl u. Kohle*, 1951, **4**, 288–90.—A new cutting oil consisting of the Na salt of mepasin sulphamide acetic acid (mepasin = saturated product obtained by hydrogenation of kogasin yielded by the Fischer–Tropsch process). This contains an active polar constituent which forms a mol layer on metal and acts as rust preventative. Hö is used as aq emulsion (0·5 to 5%), very hard waters should be preliminarily softened, and NaCl content should be $\geq 0{\cdot}05\%$. Types of work for which Hö is suitable are listed, and relation of cutting speed to concn and age of emulsion is shown. Material is also a good emulsifier for mineral oil, and Hö/oil/water emulsions can be used for work where lubricating power of aq emulsion of Hö alone is inadequate. Hö has pH 7 to 8. V. B.

1958. Where are we going with fly resistance? G. C. Decker and W. N. Bruce. *Soap Sanitary Chem.*, June 1951, **27** (6), 139.—The problem of developing and using insecticides with the min change in the resistance characteristics of the insect is discussed, particularly in relation to household insecticides for flies. From work on D.D.T. resistance, it is considered that the evidence suggests that the cause of increasing resistance is due to an ability to metabolize or degrade D.D.T. quickly. The most important factor in the change of resistance is the contamination of the medium in which the larvæ breed. A strain which has built up resistance to one insecticide does not give resistance to another compound unless closely related in chemical type. The slowest build-up was shown by synergized pyrethrins and paraoxon. In practice, change from one insecticide to another can produce multi-resistant strains. Cases are quoted where flies are resistant to D.D.T., Dieldrin, and Lindane. The ultimate problem facing the industry is inability to control insect populations. A series of recommendations in the handling of insecticides is given, stressing the danger of using residual-type products in larvicides. N. W. G.

1959. An ultraviolet spectrophotometric quality and stability criterion for medicinal liquid paraffin. R. Schnurmann, P. M. Martin, and W. F. Maddams. *J. Pharm. & Pharmacology*, 1951, **3**, 298–301.—Spectrophotometric data of absorption in 2400 to 2800 Å region is reliable test for liq paraffin quality. Stable oils show absorption min at approx 2490 Å and max at about 2710 Å., oils having ratio of absorption intensity $2710 : 2490 > 2{\cdot}0$ withstand daylight storage for periods > six months. Tabulated results show lack of correlation between spectrophotometric findings and B.P. acid test; latter is thus considered unreliable guide to oil quality. Two groups of compounds have been chromatographically separated from liq paraffin; odour-producing bodies have rapid decrease of absorption intensity at $\lambda > 2400$ Å. V. B.

1960. The conductivity of hydrocarbon transformer oil containing water and solid conducting particles. A. W. Stannett. *Brit. J. Appl. Phys.*, 1951, **2** (4), 110.—The quality of transformer oil in service has been assessed by the D.C. resistivity test. Droplets of water in oil have been observed under the microscope and found to oscillate between two electrodes in a 300-V D.C. field, transferring charge and giving rise to a high conductivity. With higher test temp, oil dissolves more water, and the amount left suspended decreases with a consequent fall in conductivity. An inverted V-shape curve results when the logarithm of the D.C. resistivity is plotted against temp, the temp at which the max occurs rising (and the D.C. resistivity falling) as the water content increases to 83 p.p.m. The results obtained with suspensions of aluminium powder in oil suggest that solid conducting particles such as carbon or wet fibres give rise to high conductivity and a resistivity/temp characteristic parallel to (and at a lower level than) new oil. The conductivity is proportional to the square of the electric stress, the quantity of conducting matter, and inversely proportional to the visc. It is concluded that in practice, it is a reasonable assumption that an inverted V characteristic indicates the presence of water; it is, however, unsafe to assume that solid conducting particles would be indicated by a low-level resistivity curve, which could also result from weak acids due to oil oxidation. C. N. T.

1961. Stabilization of wax aerosols. B. J. Eiseman, Jr. *Soap Sanitary Chem.*, May 1951, **27** (5), 106.—A wax aerosol consists of three main constituents—finely divided wax particles, the vehicle (kerosine, etc.), and the propellent (Freon-type material), the last two being mutually compatible, and the propellent forming 75% of the blend. The major formulation difficulty is due to "creaming" of the wax. This was overcome by one of two methods:—

(*a*) By adjusting the density of the solvent and solid phases to be equal. To do this, a finely powdered dense inert material was added, calcium fluoride being the most effective, as it also improved the drying of the film, its gloss, and acted as a gel preventative.

(*b*) The material can be stabilized by using a bifunctional or polyfunctional compound—dibromosuccinic acid being illustrated.

To reduce "thermal lumping," resulting from changes of temp and wax crystal growth, two approaches were experimented with—reducing the solubility of the wax in the

solvents or using materials which were completely soluble. A range of different types of Freons was tested, and these were checked and found to have no greater corrosive properties than Freon 12. N. W. G.

Derived Chemical Products

1962. Petrochemicals—the field with a future. E. Ayres. *Oil Forum*, May 1951, **5** (5), 159.—Although the main business of the oil industry is the supply of fuels and lubricants, small proportions of the petroleum supplies have for many years been diverted to a large number of other important applications. Production of petroleum chemicals has increased from 200,000 tons in 1940 to 2,700,000 tons in 1950, and there are now more than a thousand chemicals made from petroleum.

The starting points for most of them are olefins, and the most important of these is ethylene. The average yield of ethylene in the U.S.A. probably amounts to about 1 lb/brl of crude refined, and the annual total for the oil industry is probably about 2,500,000,000 lb. In 1950, 15,000,000,000 lb of ethylene were used for chemical manufacture, and most of this was from petroleum. Ethylene not recovered for use for chemical manufacture is utilized as fuel. The conditions under which the segregation of ethylene may be regarded as economically justifiable are, as a rule, when the concentration of ethylene in the refinery gas stream is above 3%, when the volume of gas in one location is sufficient to produce more than 10,000,000 lb of ethylene annually, and when accessible markets for the produced quantities exist. The long-term outlook is for low-cost ethylene, provided capital outlay for ethylene recovery can keep pace with increasing demand.

The paper supplies a list of the major uses of ethylene and suggests that, although most of the chemicals now made from petroleum are derived from olefins, the future trend may be more towards the aromatic-based compounds. A. S.

1963. Toilet bar form synthetics. V. J. Keenan. *Soap Sanitary Chem.*, May 1951, **27** (5), 27.—The possibility of making a bar or tablet from 100% synthetic is examined using an alkyl aryl sulphonate as the detergent. It is found that the whole marketing problem revolves around the established machinery and type of product for a soap tablet. The roll mill and plodders will not normally work with a synthetic, because pressure and temperature do not melt it in the same way. In use, soap has a large binding power for water, and as a result can be left wet in a dish, whereas synthetics are poorer in this respect, and tend to liquefy on the surface and run. A suitable formulation was developed to give an all-synthetic detergent milled toilet bar containing 50% alkyl aryl sulphonate with soya lecithin as a detackifier, corn starch for hardening, and talc for "slush resistance." N. W. G.

Miscellaneous Products

1964. Detergents in sewage. Anon. *Soap Sanitary Chem.*, June 1951, **27** (6), 79.—A short review of ten papers presented to the Division of Water, Sewage and Sanitation Chemistry at the 119th meeting (Pt II) of the American Chemical Society, April 1951, at Cleveland. This symposium discussed the possible problems and reviewed results due to the increasing proportion of synthetic detergents in raw sewage. The aspects discussed cover sedimentation, emulsification, possible foaming on the bio-aerators, comparative oxidation of different chemical types, effect on slime growth, and the increasing proportion of phosphate residues resulting from built detergents. N. W. G.

1965. Soaps and synthetics—present and future. F. D. Snell and C. S. Kimball. *Soap Sanitary Chem.*, June 1951, **27** (6), 27.—An examination of the present off-take of detergents (soaps and synthetics) in the U.S.A. and possible changes in the future. Soap demand has lessened due to the capture of a proportion of the market by synthetics. This results in ample supplies of non-edible fats, but the reduction in soap production has reduced supplies of glycerine—a shortage partly met by synthetic glycerine. For synthetic detergents, the supply depends upon the benzene position. This chemical is in short supply, and this is expected to curtail the rate of expansion

of alkylate detergents. Sulphuric acid shortage will affect synthetics more than soap. Phosphate builders—the major supplementary chemical for powder detergents—are now being met by increased plant capacity. N. W. G.

1966. Hand cleaners and protectants. M. A. Lesser. *Soap Sanitary Chem.*, June 1951, **27** (6), 31.—The first part of a review on the history of industrial dermatitis, factory hygiene, and the formulation of barrier creams and hand cleaners. Industrial dermatitis accounts for 65% of occupational diseases, and the incidence is growing with an increased number of primary irritants of new chemical type. Cleanliness is stressed. In an historical review of the type of cleaners, formulation is traced from sulphonated oils to formulæ showing an increased use of synthetic detergents, particularly of the anionic type. N. W. G.

1967. Non-ionics in the dry-cleaning industry. G. E. Barker and H. J. Ranauto. *Soap Sanitary Chem.*, June 1951, **27** (6), 43.—This article discusses the use of solubilized formulations as dry cleaning aids with the power of enabling a small proportion of water to be added to the process. The value of any agent must be considered in relation to the cleaning (with spotting at a minimum), the whiteness retention of the goods, odour, and emulsification properties. A number of formulations, based on proprietary non-ionics, are shown. N. W. G.

1968. Hard surface cleaners. J. C. Harris. *Soap Sanitary Chem.*, June 1951, **27** (6), 49.—This article is Pt 1 of this subject, and deals mainly with the factors responsible for the adhesion of dirt to hard surfaces, such as metal, glass, ceramics, etc. The factors quoted are: free surface energy, electrostatic charge, "ion exchange" capacity, chemical reactivity of surface, surface area, roughness of surface. A brief description of the laboratory technique of panel washing is included. N. W. G.

1969. Auto cleaning products. Pt II. M. A. Lesser. *Soap Sanitary Chem.*, May 1951, **27** (5), 31.—A review of the types of materials used for cleaning automobiles, dealing principally with washing and polishing compounds for enamel finishes—from the simple lustre oil-type polishes, through mild abrasive materials, cleaner-polishes, wax polishes, pre-waxing cleaners, aerosol wax sprays, and silicone polishes. In wax-containing polishes, carnauba wax is the most important ingredient, with various amounts of substitution by synthetic and paraffin waxes. Metal polishes, window cleaners, anti-fogging treatments, and upholstery cleaners are briefly covered. N. W. G.

1970. Emulsifier trend: sharply up. J. P. Sanderson and F. K. Abel. *Chem. Ind. Wk*, 26.5.51, **68** (19), 19.—This report reviews the American emulsifier market and estimates the 1951 consumption at 300 million lb. This is arrived at by examining the market by industries, and by chemical types of emulsifiers. The largest using industry is synthetic rubber and resin polymerization at 120 million, the bulk of this (115M) being used for emulsion polymerization of GR-S, the remainder being mainly for vinyl, styrene, and acrylic resins. The food industry (80M) uses mainly esters with glyceryl monostearate (59M) as the chief emulsifier. The other listed items are polishes (35M), cutting oils (25M), cosmetics, toiletries, and pharmaceuticals (10M), dry-cleaning soaps (8M), textile and leather (7M), insecticides (6M).

Subdividing by chemical types, soaps (168M) are the largest group; sodium soaps (60M), rosin soaps (58M), covering the largest volume. These are used in all industries except food and insecticide groups. Fatty acid esters (92M) have glycerine derivatives (62M) and polyoxyethylene (18M), with uses in cosmetic, food, and insecticide classes. The group of sulphonated oils (35M) has petroleum sulphonates (19M) as the main emulsifier, and is used chiefly in cutting oils, dry-cleaning soaps, and textiles and leather. The miscellaneous emulsifiers (5M) include amides, lecithins, lignins, polypeptides, etc. N. W. G.

1971. Accelerated weathering of paint and varnish films. P. J. Gay. *J. Oil Col. Chem. Assoc.*, Feb. 1951, **34** (368), 43.—A brief historical survey of the development of accelerated weathering is given. The advantages and disadvantages of the accepted methods of testing are discussed.

The accelerated weathering apparatus at the paint research station prior to and later than 1947 is described. The apparatus, rebuilt in 1947, enables the variation of, the time and intensity of ultra-violet light radiation, amount of water treatment, and period of revolution of the test drum. Paints and varnishes tested and the results obtained are tabulated. Paint films treated with oxygen at elevated pressures, to accelerate its diffusion into the films, broke down in a manner comparable with that obtained outdoors. It is thought that a study of the oxygen concentration variable will provide one of the most useful developments in durability testing procedure for paints. Details of standard accelerated weathering procedure are appended, together with photographs of breakdown under different accelerated weathering cycles. There are seventy-eight literature references. D. K.

1972. Changes in paint films. A. V. Pamfilov and E. G. Ivancheva. *Paint Technol.*, May 1951, **16** (185), 193.—Physical characteristics of paint films examined were tensile strength and relative elongation. Boiled linseed oil and a series of pigments of the metal-oxide type were used as test materials. Incorporation of a pigment in an oil film markedly increased the mechanical strength, increase in tensile strength was greater when the pigment reacted chemically with the binding medium. The effect of different pigments varied considerably, films containing reactive pigments aged to a greater extent than those containing unreactive pigments. Results of tests carried out on films exposed indoors, at 70° C, and in the open air are discussed. Analytical procedures used included chloroform extraction. Results showed that only in the case of litharge fairly large quantities of linoleates were formed. All pigments reacted to some extent with boiled linseed oil after paints had been applied in thin layers to a glass surface.

Reactivity of pigments in films containing partially saponified linseed oil and in films prepared from linseed oil fatty acids are discussed. Mechanical properties of linseed oil fatty acid paints deteriorate fairly rapidly, especially under outdoor-exposure conditions.

The reaction between pigment and oil in the bulk of the paint differs from that in the film. The properties of the film are mainly governed by those of the products of the pigment–oil reaction. There are seventeen literature references. D. K.

1973. Plasticizer action. P. Piganiol. *Bull. Ass. franç. Tech. Pétrole*, 1951, (87), 35–49.—Main plasticizer requirements are considered; compatibility, mechanical properties of resin/plasticizer complex, electrical properties. Recent theoretical studies of plasticizing action are reviewed, with particular reference to the polymer–plasticizer interaction constant (μ), which can provide a quant indication of compatibility. Curves are given for a phenolic plasticizer and polyvinyl chloride tested in an instrument (" ephimeter ") which simultaneously measures internal friction and the modulus of elasticity. V. B.

1974. Sonic agglomeration of carbon black aerosols. C. A. Stokes. *Chem. Engng Prog.*, Aug. 1950, **46** (8), 423–32.—Methods of production of carbon black are briefly summarized, and the characteristics of carbon black aerosols and existing methods for their agglomeration are noted. Details are presented of large-scale experimentation, using actual commercial conditions, with sonic energy as a means of agglomerating carbon black. The apparatus and procedure employed are described in detail, and the results of collection in this manner are evaluated as being equivalent to those obtained with the Cottrell electrostatic system. J. G. H.

1975. GR-S X : Is it a development of the " oil technique "? H. C. Harrison. *India Rubber J.*, 1951, **120** (20), 786.—Information now available indicates that GR-S X is 75% cold polymer and 25% mineral oil, the product being tough and highly viscous. The process appears to be a development of the " oil technique " discovered by the author in England during the war. His process involved the soaking of thin sheeted GR-S in mineral oil overnight, and subsequent mixing with other ingredients on the mill. It was shown that GR-S would absorb up to 50% wt oil, but 20 to 25% was the amount generally allowed. The properties of this material are described, and the obvious advantage of adding the oil to the emulsion or latex at an early stage in the process is pointed out. C. N. T.

CORROSION

1976. Corrosion and its control. M. E. Parker. *Oil Gas J.*, 3.5.51, **49** (52), 131.—Pt 9 of this series deals with cathodic protection of tank bottoms. Two methods are available, rectifiers and ground beds, or expendable anodes; and choice depends on several factors. A typical rectifier installation consists of 40- to 100-amp units distributed about the tank farm, each rectifier giving protection to from four to six 55,000-brl tanks.

A magnesium anode system consists of a separate group of anodes in a concentric ring around each tank to be protected, as many as twelve to twenty anodes may be connected together and then to the tank through a current-measuring shunt and limiting resistor.

Current requirements for tank bottoms range from 2 mA/sq. ft. to as low as 0·25 mA/sq. ft. G. A. C.

1977. Corrosion and its control. M. E. Parker. *Oil Gas J.*, 10.5.51, **50** (1), 144.—Pt 10 of this series deals with economic comparisons between total operating costs of similar structures with and without cathodic protection.

Three general types of comparison are considered, and include side-by-side, off-and-on, and fresh-start comparisons. G. A. C.

1978. Corrosion in amine gas treating plants. F. C. Reisenfeld and C. L. Hughes. *Refiner*, 1951, **30** (2), 97–106 (*Amer. Soc. Mech. Engrs, New Orleans*, Sept. 1950).—The increased domestic demand for natural gas has led to the development of gas-purification and dehydration processes. Amine processes, which have been carried out in carbon steel, have proved to be corrosive. Detailed notes are presented on a programme designed to find alternative materials of construction to eliminate or retard the corrosion. In glycol–amine plants, solution temp in hottest pass of heat exchangers, degree of saturation of the solution, and elimination of oxygen have proved to be important factors, while for aqueous amine systems the reboiler temp and the presence of oxygen and oxidation products are important. Alternative materials of construction suggested are aluminium 35 and 4–6 Cr ½ mol steel for glycol–amine plants, and 304 and 316 stainless steel, Monel, Inconel, and Carpenter 20 steel for aqueous amine systems. R. W. G.

1979. Corrosion-prevention program for T.C.C. gas plant. C. A. Murray and M. A. Furth. *Oil Gas J.*, 24.5.51, **50** (3), 112.—History of severe corrosion and corrective measures taken at unit of Pure Oil Co. Smith Bluff refinery, Nederland, Texas, are given. Examples illustrate failures and replacements necessary in tubular heat-exchange equipment; and a summary of a chemical survey shows types of corrosives and amount of water which produced the corrosion in the system. Principal agents were ammonia, hydrogen sulphide, and ammonium salts of organic acids.

Corrosion-preventive measures include removal of water with coalescers at appropriate points in gas plant.

Benefits to date of the prevention methods are discussed. G. A. C.

1980. How hydrogen attacks and damages steel in refinery equipment. R. T. Effinger, M. R. Renquist, A. G. Wachter, and J. G. Wilson. *Oil Gas J.*, 17.5.51, **50** (2), 99.—The mechanics of hydrogen penetration and rate of diffusion are dealt with. Effects of penetration include blistering, fissuring, and loss of ductility.

Tests were made of the effects of hydrogen damage on a blistered vessel, by application of pressure until rupture occurred. A survey of plant-operating conditions on a catalytic cracking gas plant is given. Methods of investigating the environments which cause hydrogen attack include chemical analyses of process streams, use of plant itself in which processing conditions can be varied, and synthesis of processing streams in which one component at a time can be varied. Membrane types of hydrogen-accumulating devices were developed for measuring activity of environments promoting hydrogen penetration; and determination and effect of pH studied. It was found that concentrations of cyanide salts caused a marked effect. Three hydrogen-active environments in plants are recognized and control systems considered. Seven figures illustrate the article, and thirty-two references are given to this important work. G. A. C.

1981. Hydrogen attacks of steel in refinery equipment. R. T. Effinger, M. L. Renquist, A. Wachter, and J. G. Wilson. *Refiner*, 1951, **30** (5), 132–44.—This discussion deals with low-temp hydrogen attack of low-carbon steel, commonly used in refinery construction, associated with aqueous corrosion, in which hydrogen which penetrates the steel is a product of the electrolytic corrosion cell. This discussion is divided into the following sections: (1) mechanism of hydrogen penetration, (2) effects of hydrogen penetration on steel, (3) survey of plant-operating conditions, (4) instruments and testing methods, (5) laboratory investigations, and (6) avoidance of hydrogen damage in the plant. The control of the hydrogen attack may largely be affected by pH control and washing to remove undesired salts. Thirty-two literature references.

R. W. G.

1982. Importance of metallurgical factors in corrosion phenomena observed in the petroleum industry. M. Pruna, B. Le Boucher, and P. Lacombe. *Rev. Inst. franç. Pétrole*, 1951, **5**, 145–55.—Examples of corrosion of four metals commonly used in refinery construction are considered—ordinary steel, brass, cupronickel, stainless steel. Particular attention is paid to the influence of impurities in the metal (S inclusion) in the case of mild steels (0·15 to 0·2% C), of thermal treatments (welds), and of stresses. Importance of microscopic investigation is underlined (thirty-six photomicrographs reproduced), and likewise the careful preservation and examination of corroded material, as a guide to accurate assessment of the corroding conditions.

V. B.

1983. Corrosion-resistant materials. L. W. Johnson and E. J. Bradbury. *Trans. Inst. mar. Engrs*, Apr. 1951, **63** (4), 59.—Corrosion and erosion remain the principal causes of maintenance costs in marine-engineering installations, and the purpose of this paper is to collect useful information on the application of corrosion-resistant materials in marine engineering and to present it in a compact form which will be of use to builders and users of marine-engineering equipment.

The paper consists of three main sections:—

(*a*) A brief introduction to the theoretical aspects of corrosion.

(*b*) A description of the corrosive media normally encountered in marine engineering.

(*c*) A list of the general corrosion-resisting properties of the metals and alloys commonly used in these applications.

It is concluded that corrosion problems encountered in marine engineering vary considerably from installation to installation, but there are available to marine engineers materials resistant to practically all conditions of service. However, the use of a resistant material is not the answer to all problems, because in many cases a change to such a material would not be an economic proposition. Nevertheless, it is frequently possible to arrange for a compromise by which, by small re-design of equipment and the adoption of an alloy slightly more resistant to the service conditions than that previously used, an economic answer can be obtained.

Good design remains one of the principal factors in overcoming corrosion and erosion problems, but this must always be accompanied by the correct selection of materials of construction.

A. S.

1984. Protection of well casing against external corrosion. R. E. Hammond and S. P. Ewing. *World Oil*, Feb. 1951, **132** (2), 154.—Casing leaks in the Loudon Pool, Ill., were traced to the Tar Springs formation at a depth of about 1200 ft. Research into the causes of these leaks revealed that 90% of the corrosion was electrolytic; the use of insulating connexions between casing and flow lines proved effective in reducing the leaks in the casing, but greater corrosion of surface lines was experienced.

The design of the cathodic-protection system and general problems are outlined.

A. J. H.

1985. Practical and relative simple corrosion tests found effective. F. L. La Que. *Refiner*, 1951, **30** (2), 120–3.—A description of tests on various materials for corrosion resistance, which may be conveniently carried out in plant under operating conditions. This method is based on the ASTM Recommended Practice for Conducting Plant Corrosion Tests, A224–41. This procedure of testing is often to be favoured over laboratory methods, when it is difficult to establish identical conditions. R. W. G.

1986. Current requirements for cathodic protection. M. E. Parker. *Oil Gas J.*, 7.6.51, **50** (5), 85.—Pt 14 of this series concerns neutralization of positive charges produced by hydrogen ions; current required will be low for places where the neutralized ions will tend to form a film; and high where film is removed as fast as formed.

Pipelines in soil will require 1 to 2 mA/sq. ft. of exposed surface, bare steel tank bottoms have similar requirements. Steel in seawater requires 20 to 30 mA/sq. ft.

G. A. C.

1987. Current requirements by field test. M. E. Parker. *Oil Gas J.*, 14.6.51, **50** (6), 109.—Pt 15 in this series discussed a test site of scrap 2-inch pipe to duplicate conditions at the rectifier installation in order to estimate current requirements; location, current, and duration of test being considered.

The effect of combining several units along the line must be taken into account.

G. A. C.

1988. Full protection of coated lines with magnesium. M. E. Parker. *Oil Gas J.*, 31.5.51, **50** (4), 119.—Part 13 of this series reviews advantages and disadvantages of the magnesium-anode method of protection, and application of complete protection to coated pipelines discussed.

After initial installation of part of estimated quantity of magnesium, a period of three weeks or more will be necessary to allow time for complete polarization of the line.

A protective potential of 0·85 V is required. Connexion to pipe itself may be made by thermite process; no mechanical connectors should be used. G. A. C.

1989. Pipe line corrosion. G. G. Portch. *Electr. Times*, 1951, **120**, 4–10.—Principles underlying corrosion and cathodic protection are outlined. Buried sacrificial anodes (*e.g.*, 22-lb Mg blocks) connected to the pipe can be used, but protection by applied D.C. is preferable. Since protective current will vary with changes in soil resistance (*e.g.*, due to rain) constant current equipment is desirable. Brief illustrated description is given of such units as fitted every 5 to 10 miles along 562-mile Kirkuk–Mediterranean pipeline. Oil-immersed transformer combined with Se rectifier supplies 40 amp at 50 V. Ground bed is usually within 700 yd of pipe, potential of which is maintained at −0·8 V relative to soil; to measure this accurately special $Cu/CuSO_4$ half-cell is required, illustrated description of which is given. V. B.

1990. Magnesium anodes for "hot spot" protection. M. E. Parker. *Oil Gas J.*, 17.5.51, **50** (2), 136.—Pt 11 of this series deals with provision of additional current to compensate for "end losses."

A table gives amount of pipe of various sizes which can be afforded the desired drainage by different size anodes, based on 50% anode efficiency.

A graph shows current output and anode life as determined by soil resistivity.

G. A. C.

1991. Corrosion control in water flooding. F. Latter. *World Oil*, Jan. 1951, **132** (1), 141.—A treatment involving the use of a mixture of a quaternary ammonium compound, a condensed ethylene oxide–fatty acid wetting agent, and sodium nitrite is recommended for controlling corrosion and micro-biological growths, and is stated also to increase the intake rates of wells.

Results of tests are given graphically. References. A. J. H.

Engines and Automotive Equipment

1992. Radial engine with variable compression and no master rod. G. S. Hoell. *Auto Ind.*, 1.12.50, **103** (11), 45.—A description of the Sharpe aero engine, evolved to make the fullest use of high-octane fuel. The C.R. of the cylinders can be varied during flight so as to operate at an optimum rate of fuel consumption, thus facilitating an increase of cruising radius with a given load of fuel. J. G. H.

1993. Recent developments in the R-4360 engine. E. A. Ryder. *S.A.E. Trans.*, Oct. 1950, **4** (4), 559–70.—Problems involved in the development of large aircraft engines are discussed, and the development of the Wasp Major engine is described in some detail. Difficulties encountered in the disposal of waste heat and the methods adopted for their solution are outlined, together with the elimination of troubles due to lead deposits. J. G. H.

1994. Studebaker's new engine. J. Geschelin. *Auto Ind.*, 1.11.50, **103** (9), 38.—Details are presented of a new overhead-valve V-8, developing 120 h.p. with a piston displacement of 233 cu. in. Present C.R. is 7·1, but basic design is such as to allow for increased C.R. without appreciable change. J. G. H.

1995. Some aspects of fuel injection research in England. W. E. W. Nicolls. *Voodr Koninklijk Inst. Van Ingenieurs (Holland)*, Jan. 1951, pp. 93–113.—Research aims are enumerated and discussed with respect to : (1) the influence of the many variables on the performance of injection apparatus, (2) a better appreciation of the requirements of the apparatus for which equipment is intended, (3) developments to improve the reliability and life of the equipment. Measurement of atomization is described, and the jerk-pump apparatus and results of atomization experiments are shown. The injection-rate analyser is described and is shown. High-speed photography of fuel sprays and filtration requirements are described and illustrated. Photomicrographs of alumina particles used in filter and wear tests, and graphs of the solid material retained by the filter, are shown. The indicating apparatus is illustrated, and a typical diagram is shown. Engine-starting investigations on the functioning of the Pintle and Pintaux nozzles are described. A three-page discussion on this paper is included. W. H. C.

1996. Increased efficiency for diesels provided by sealing rings. Anon. *Refiner*, 1951, **30** (2), 113–14.—The use of gap-sealing piston-rings gives increased power and reduced fuel and lubricating-oil consumption, resulting in fewer overhauls, compared with ordinary rings. Figures demonstrating these points are given for cyls of different degrees of wear. R. W. G.

1997. Gas turbine propulsion for ground vehicles. W. M. Brown. *S.A.E. Trans.*, Jan. 1951, **5** (1), 81–93.—Operating experience obtained with a Boeing turbine installed in a Kenworth chassis is utilized in assessing the prospects of gas-turbine propulsion for road haulage. From the data obtained it is concluded that while there are possibilities for the gas turbine in this direction, it remains to be proved, and has not yet reached its max in performance and development. It is not anticipated that its cost will be prohibitive. J. G. H.

1998. Combating ice in gas turbines. Anon. *Flight*, 1951, **59** (2203), 414–15.—Three patented systems for overcoming the problem of ice formation on the air intake and adjacent components of aircraft gas-turbine units are described and illustrated diagrammatically. Although differing considerably in detail, all three systems are generally similar in principle, air being tapped from the compressor and delivered to a conveniently sited auxiliary combustion chamber which is separate from the main combustion system ; the hot gases generated in the auxiliary chamber are distributed to appropriate points for anti-icing purposes. N. L. A.

1999. Truck road performance—actual vs computed. C. Saal. *S.A.E. Trans.*, Jan. 1951, **5** (1), 18–25, 93.—The method of computing truck road performance developed by Stamm and Lamb is examined from the standpoint of accuracy. The method is demonstrated to be sound, and it is pointed out that the use of the basic factors now available for the resistances to motion can be expected to result in an average difference between computed and actual performance of 5 to 10% of computed performance ; also actual performance is always greater than computed figure for operation on the level, and is generally less than that computed for grades of 3% and steeper. J. G. H.

2000. Predicting road performance of commercial vehicles. A. F. Stamm and E. P. Lamb. *S.A.E. Trans.*, Apr. 1950, **4** (2), 147–60.—Previous formulæ evolved for performance calculations are recorded, and the necessity for taking into account air resist-

ance is emphasized. The fundamental considerations involved in the assessment of vehicle performance are reviewed, and a method is outlined for evaluating gradient-climbing ability at a given or assumed road or engine speed, based on the use of a series of charts and tables covering road speed, engine r.p.m., rolling and air-resistance horsepower, and chassis frictional horsepower. Instructions are given for the use of these media, and max attainable road speed with given equipment together with optimum gear ratio under given conditions are discussed. J. G. H.

2001. Brake torque. G. A. G. Fazekas. *Auto Engr*, May, 1951, **41** (540), 185.—The torque output of a brake invariably fluctuates, even if the nominal actuation load is constant. For this reason, nearly all tests aim at determining average torque output. Neither the method nor the interpretation of such tests is, as yet, standardized, and the purpose of the paper is to consider some of the fundamentals involved.

Although the whole subject is treated in relation to motor-vehicle brakes, the conclusions reached are also applicable to aircraft or industrial brakes. Brake tests carried out are described, together with method of brake actuation, and the method of interpretation of the results is given.

Appendix I shows how the response of brake systems is calculated, and Appendix II shows how the average torque output with regard to time differs from that referring to distance. A. S.

2002. Modern carbon composites. S. A. Murray. *Auto Engr*, May 1951, **41** (540), 182.—In recent years fabricated carbon and metal carbon composites have become key materials in a wide variety of engineering applications. The major properties of carbon from a mechanical viewpoint are: (1) self-lubricating; (2) high resistance to temp and thermal shock; (3) availability in impervious form; (4) machineability to a high degree of accuracy; (5) it is not wetted by molten metals; (6) high resistance to wear.

As carbon has no liquid phase, it cannot be cast like metals, and carbon articles are usually fabricated from powder, agglomerated with a suitable binder. After compressing the mixture under very high pressures to the desired shapes, the compact is subjected to a lengthy heat treatment in a protective atmosphere at very high temp.

Regarding specific applications, primarily of the metal carbons, the following are of interest to engineers in the automobile and allied industries: (1) clutch-release bearings; (2) plain bearings; (3) pump and shaft fluid seals.

In addition to these, carbon, metal-carbon composites, and sintered metals of specially developed compositions are extensively employed elsewhere. Some of these applications are listed. A. S.

2003. Application of rocket power to aircraft. H. R. Moles. *Aero Engng Review*, 1951, **10** (4), 24–30.—The application of rocket power to past, present, and future types of aircraft is described, consideration being given only to those types of rocket power-plant employing liquid propellants. The more common propellants for aircraft application are described, as well as methods of storing liquid oxygen. The advantages of rocket power-plants for powering aircraft are listed, and a description given of the arrangement of a typical pump-fed rocket power-plant. The possibilities of building an effective rocket-powered interceptor aircraft at the present time are also examined. N. L. A.

Safety Precautions

2004. Safety for working inside petroleum tanks. J. Z. Walczynski. *Nafta*, Jan.–Feb. 1950, **6**, 26–32; Mar. 1950, **6**, 63–7. (From the work of the Forensic Medicine Institute of Cracow University.)—Quoting German, Russian, British, and Polish books and instructions, the author discusses toxicity of petroleum products and fractions. He goes on to consider the work involving danger of poisoning by breathing-in paraffins, cyclic compounds, sulphur, and nitrogen compounds, such as may be found in petroleum, and by absorption of them through the skin.

Most experienced workers go into fresh air immediately they feel first signs of poisoning. All stages of poisoning, including those likely to cause permanent injury, are

described. The precautions necessary to avoid fatal accidents are fully discussed. Thorough emptying and steaming or repeatedly filling the tank with hot water is suggested. One must bear in mind the possibility of formation of an explosive mixture if there is any welding to be done. For toxicity tests animals could be used, and cats are found to be most sensitive. Explosive limits can be tested, using sampling bottles or continuous-flow instruments. All tests must be done immediately before the entry of men, because changes of humidity or temp will affect internal conditions in tanks. Should it not be necessary to carry out any welding, all accidental sparking from nails, tools, or electric discharges must be eliminated. Tanks must be earthed, and safety electric lamps must be used, preferably fixed on the workman's head. To protect the respiratory organs gas masks must be used, but if percentage of oxygen is less than 16 to 17 absorbent types must not be worn. Breathing apparatus is recommended. Rescue belts of material resistant to petrol vapours and special clothing are recommended. Medical staff should have the final say in matters of choice of workers for such jobs, who should also be examined immediately before beginning work. They should be taught about the danger of their work. First aid and rescue preparations, planning of work, and shift assignment are described, and a rota suggested for seven men, of whom only two would enter the tank. Safety regulations are in most countries backed by the criminal code, since their disregard can easily cause death. M. S.

2005. Present status of air-pollution research. H. P. Munger. *Mech. World*, May 1951, **73** (5), 405.—Methods of reducing air pollution have been studied for many years, and, during the past two years, many valuable symposiums have been held and bibliographies compiled on contaminants in the air and how to remove them. Although a tremendous amount of information has been accumulated, there is still a large amount unknown about the control of air pollution. More economic methods of separating contaminants from the air must be developed, and medical research on the allowable limits for single and mixed contaminant systems is especially needed.

From the information available, it is obvious that each plant presents an individual research problem, and the method for reducing contaminants must be adjusted to fit this individual situation.

A great deal of information is available as a guide in sampling and analysing air contaminants, and the newer techniques make possible the determination of the concentration of air contaminants at various altitudes, as well as at ground level. Close co-ordination between the air-pollution control officers, industry, and research organizations is required for the most rapid progress and, by co-operative effort, the air will gradually improve as additional equipment is perfected and used. A. S.

BOOK REVIEWS

Active Carbon. J. W. Hassler. New York: Chemical Publishing Co., Inc., 1951. Pp. v + 384.

The author states that the "text has been prepared for operators and research workers in industry," and to these people the book is to be particularly recommended. It is, however, far more than a guide for industrial workers, for it contains adequate literature reviews of almost every application of activated carbon, and as such is of considerable value to all workers employing this material.

The book is divided into three parts. The first part deals with the preparation of properties of activated carbon, the second part with the application of activated carbon to various industrial processes, and the third part with experimental methods employing activated carbon in laboratory studies.

Each chapter is a separate essay reviewing both the scientific and patent literature with the author's opinions based upon his considerable experience with the subject under consideration. The chapters may be read individually by people who are specifically interested in one aspect of the subject, so that the book forms a useful reference text for anyone searching for a particular application of activated carbon. The whole book is delightfully readable, and will find a place in the library of a variety of workers.

The chapter on the theories of absorption covers the various theories of absorption at solid and liquid surfaces, and discusses in particular the theories of chemical and physical absorption. In addition to references to standard text-books on the subject, this chapter contains some thirty-eight references to original articles. A further chapter which merits special mention is that dealing with the recovery of solvent and other vapours. Here again the literature is thoroughly reviewed, and the cost of operating activated-carbon systems compared with those of oil stripping. The chapter dealing with the separation of gases is of interest to the petroleum industry, although the "Hypersorption process" finds only very brief mention.

The book is attractively prepared and printed, free from typographical errors, and is to be recommended highly as a readable exposition of a fascinating subject.

F. M.

Der Petroleum-Ingenieur. H. Umstätter. Berlin: Springer-Verlag, 1951. Pp. xii + 552. DM 46.50.

This book, the author states, has been prepared as a text-book and a reference book for workers engaged in the petroleum industry. The text is divided into three main parts, the first dealing with production, including the geography and geology of petroleum, exploration and exploitation, storage and transport, and research on the classification of crude petroleum. The second part deals with refining, and is divided into physical processes, including distillation, crystallization, filtration, and sedimentation, into physical chemical processes, including adsorption, extraction, sweetening, and the use of inhibitors, and into chemical processes, including cracking, reforming, solvent extraction, polymerization, oxidation, etc. The third part deals with the application of petroleum products, and includes chapters on motor fuels, lubricating oils, fats, waxes, paraffins, phenols, naphthenic acids, bitumen, asphalts, briquettes, and insecticidal materials. To cover such a wide range of subjects within the compass of a single text-book has naturally resulted in certain sections being covered only in brief outline. Thus the initial part dealing with the production of petroleum is an introduction to the subject rather than a treatise.

Laboratory procedures are dealt with in considerable detail, and numerous nomographs and tables are included to facilitate design calculations and conversions. The section on solvent extraction and on viscosity determinations is particularly good. Specifications of various products covering most European countries are given in tabular form, the chapter on transformer and insulating oils, for instance, giving details of test procedures in England, America, Switzerland, France, Sweden, and Italy and the specifications effective in these countries. The details of refinery operations are illustrated by flow diagrams, and a separate chapter is included to cover instrumentation and automatic control principles.

It is, however, surprising that a text published in 1951 should contain no reference to catalytic cracking. The book may be recommended for students who wish to combine with their studies of petroleum technology a study of the German language.

F. M.

ADDITION TO THE LIBRARY

La Mineria en Colombia. El Petroleo. Banco de la Republica, Boletin Grafico No. 11, Bogota, December 1950, pp. 31.

This booklet includes historical, legal, and geological aspects of the petroleum industry in Colombia. Approximately 17% of the superficial area of Colombia has petroleum-bearing possibilities. Commencing 1907 up to 1931, eighty wells were drilled, none of which was commercially successful. From 1931 to 1950 a further 105 wells were drilled, and production in 1949 was about 30 million barrels, or 0·89% of world production. Refining capacity of Colombia is 6 million barrels per annum to meet local needs, and an additional 1·5 million barrels of refined products is imported. Maps are given showing details of the various concessions, and coloured illustrations depicting origin of petroleum, drilling, and refining are included. In 1949 petroleum-concession charges contributed 6·9% of the national income.

A. C.

CORRESPONDENCE

To the Editor, *Journal of the Institute of Petroleum.*

"PETROLEUM GEOLOGY"

Sir,

While thanking your reviewer for his kind remarks about my book "Petroleum Geology," which appeared on page 373A of the July *Journal*, may I venture to draw his attention to the fact that a brief discussion of oilfield pressures and their geological implications can in fact be found on page 53; and that he is therefore somewhat severe in chiding me for "the complete absence of any discussion" of this interesting subject.

E. N. TIRATSOO.

15 Redcliffe Gardens,
London, S.W.10.
August 22, 1951.

THE INSTITUTE OF PETROLEUM

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The current Entrance Fees and Annual Subscriptions for membership of the Institute are :

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Student Members	Nil	1 1 0

NEW MEMBERS

The following elections have been made by the Council in accordance with the By-laws.

Elections are subject to confirmation in accordance with the By-laws.

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McCulloch, C. E.
Jordan, L. T.

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Reed, J. C.
Smither, A. L.

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McGregor, H. W.
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Messent, R. L.
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Harris, A. G.
Rowland, R. P.
Hogg, G. A.
Simpson, J.
Hulme, N. E.
Slater, A. E.
Jenkins, D. F.
Wilson, K. S.
Kelley, H. H.
Wilson, R. D.

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Tennant, F. E.
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Nicholson, M. R.
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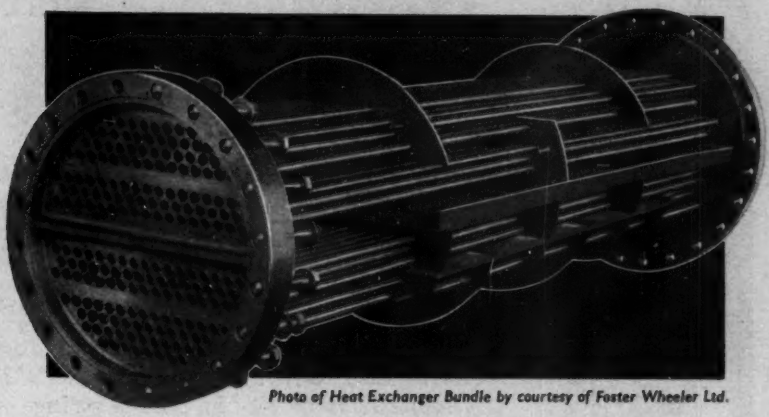

Photo of Heat Exchanger Bundle by courtesy of Foster Wheeler Ltd.

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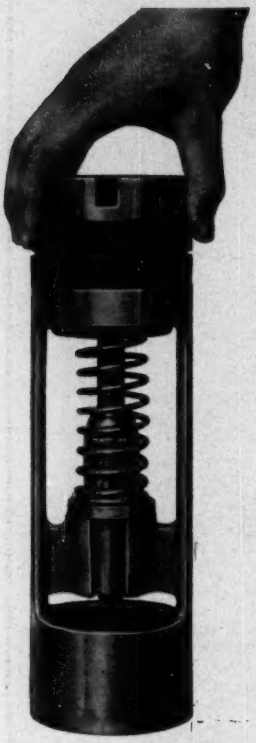

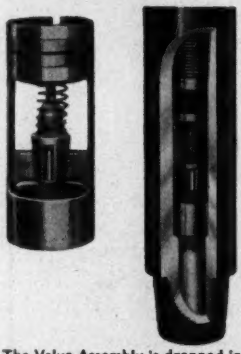

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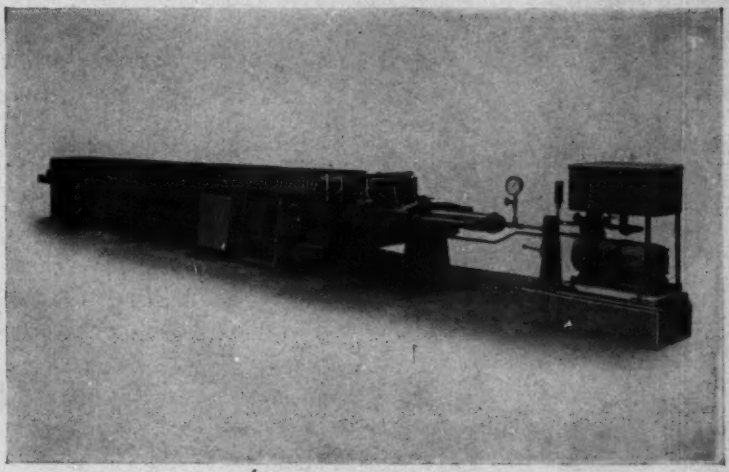

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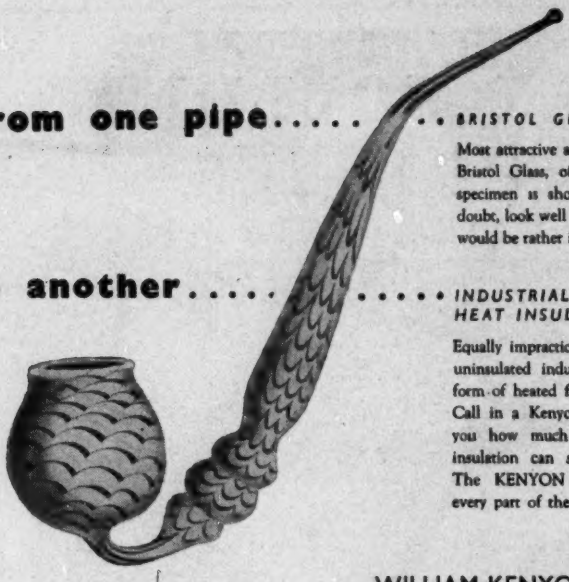

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to another.....

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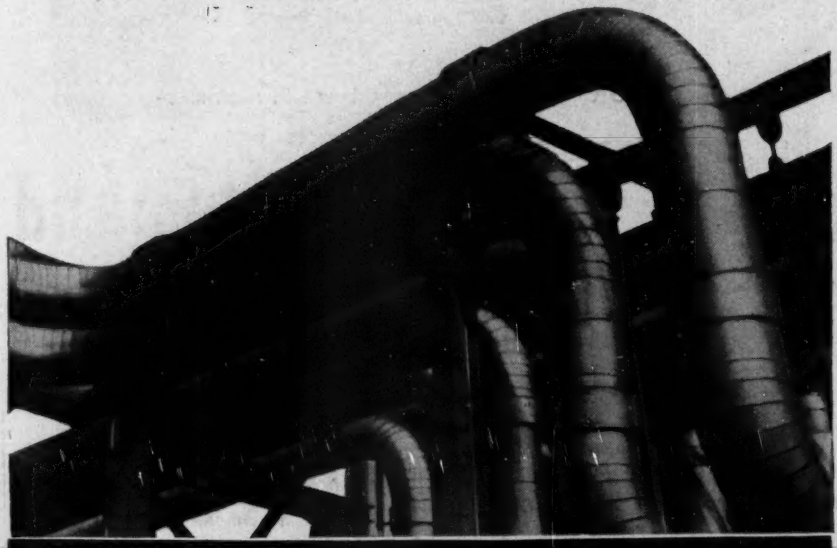

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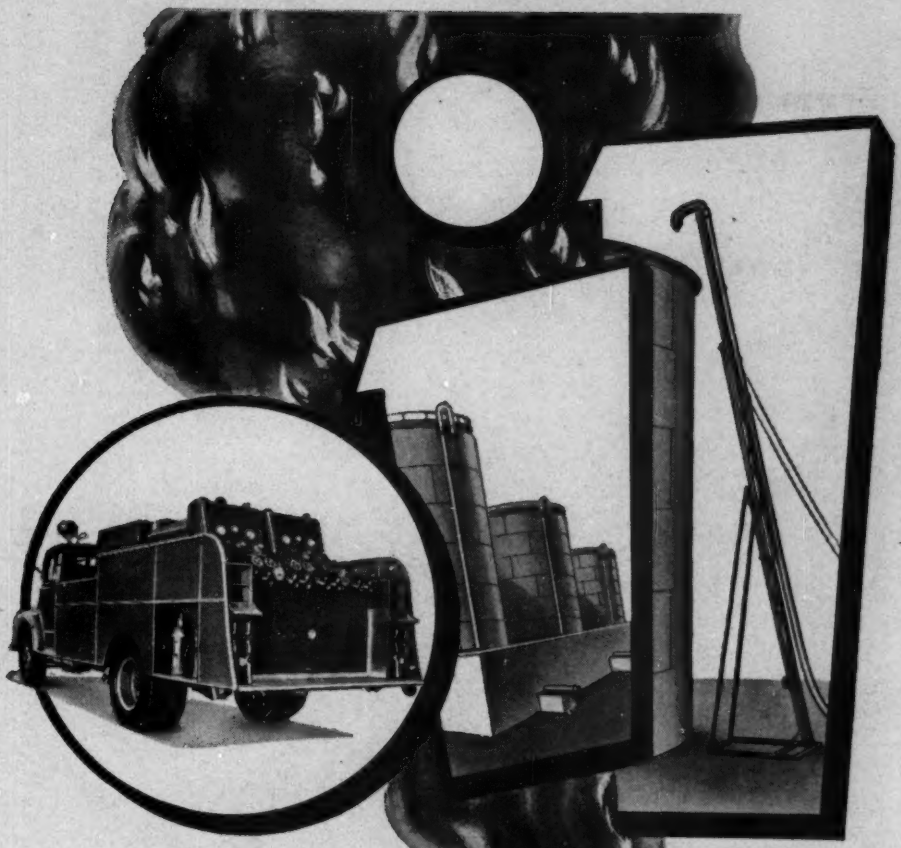

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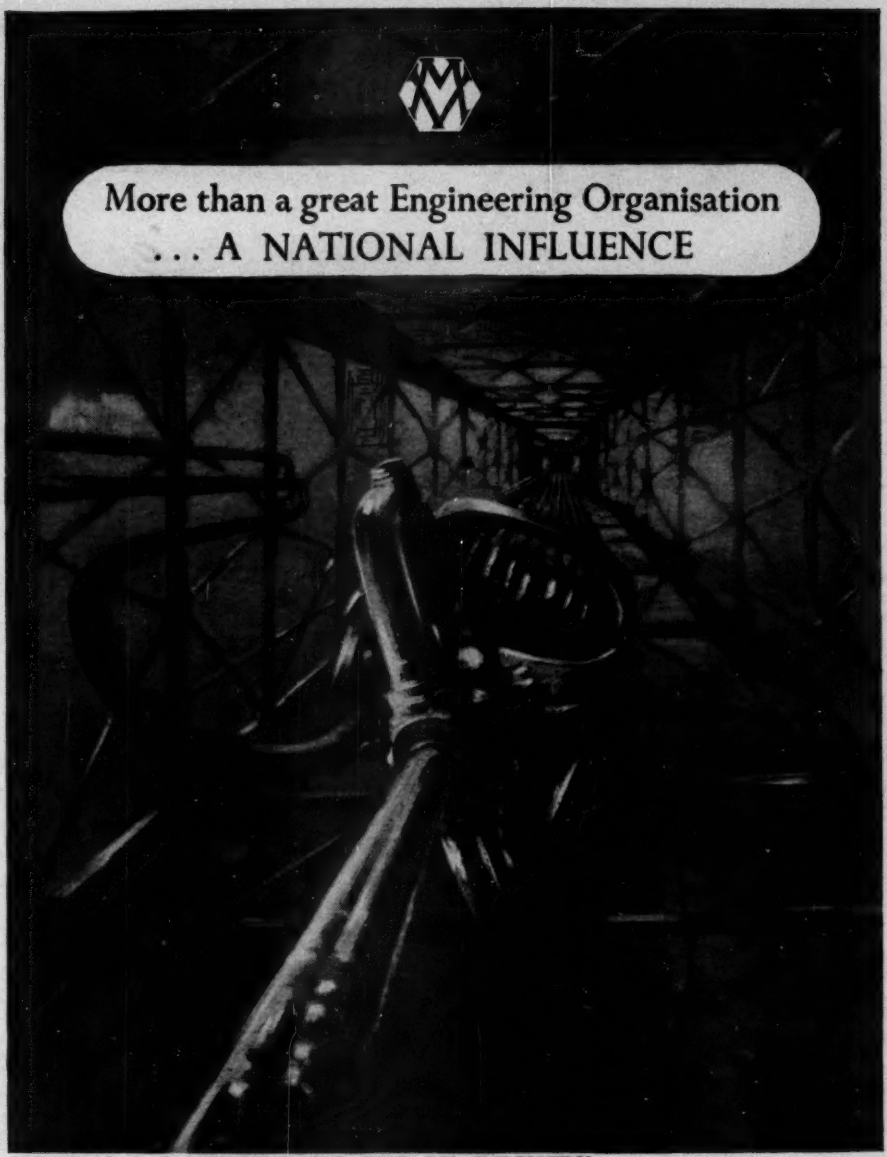

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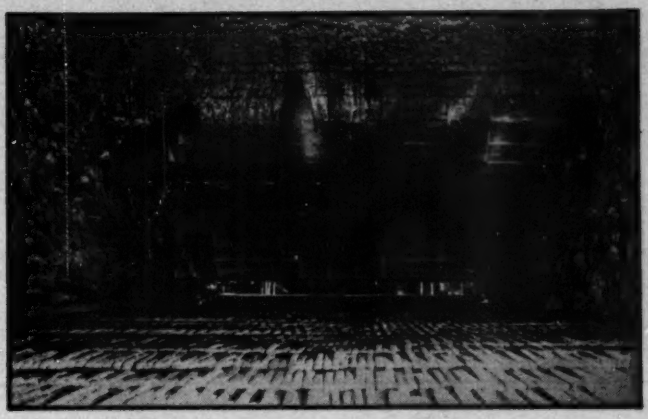

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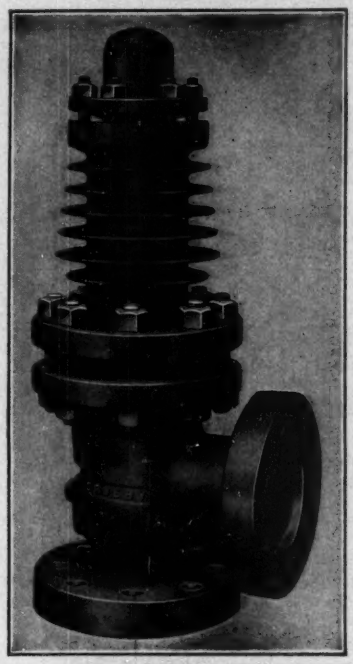

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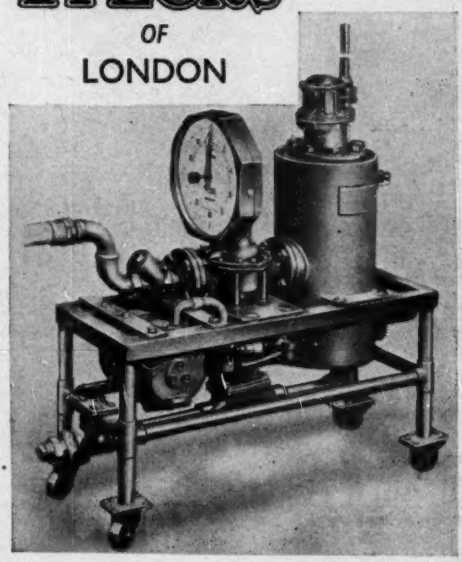

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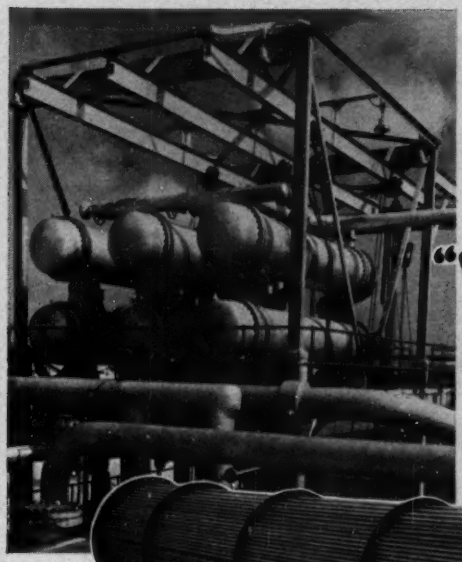

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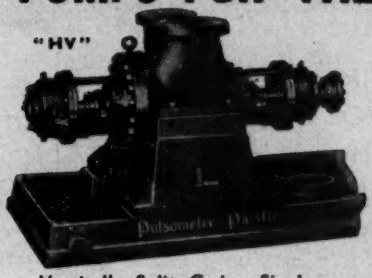

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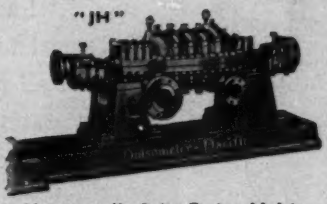

Horizontally Split Casing Multi-Stage Pumps for Hot Oil.

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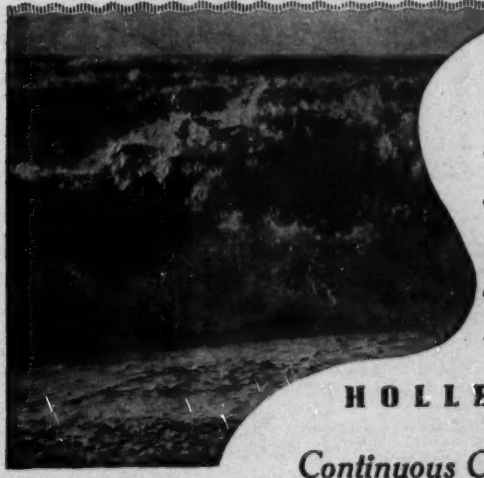

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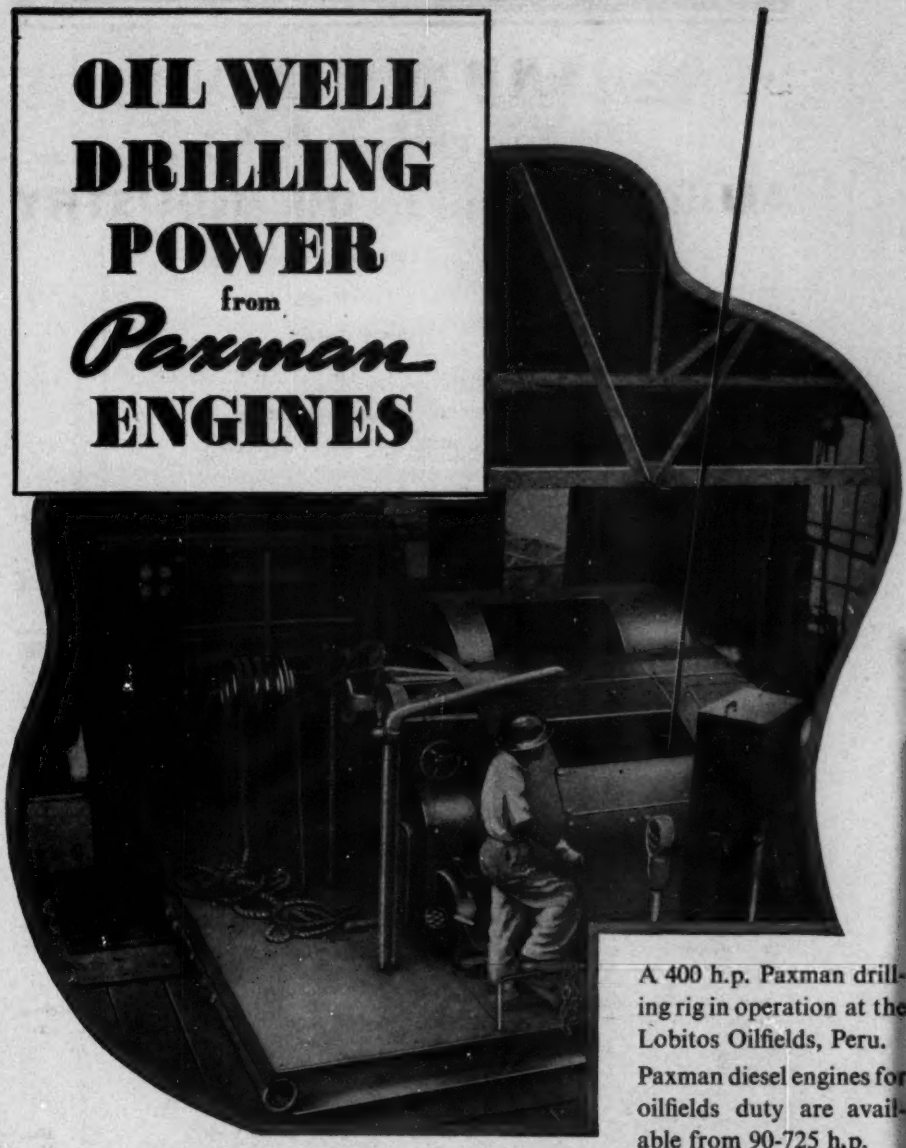
OIL WELL DRILLING POWER from *Paxman* ENGINES



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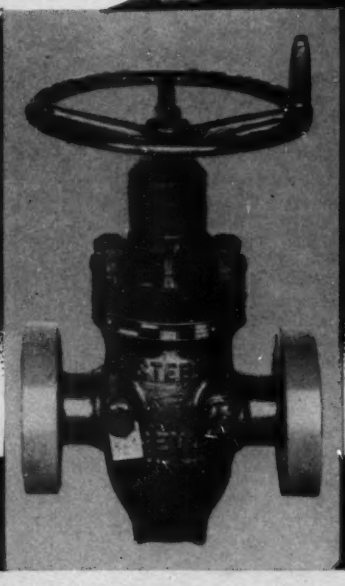

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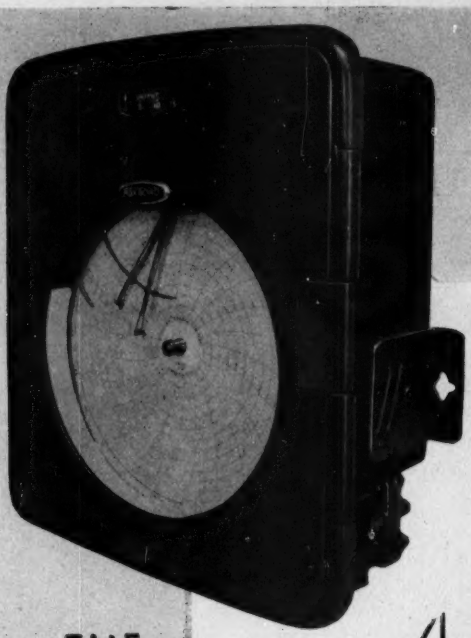

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TO THE CATHODE

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